Porous evaporators with special wettability for low-grade heat-driven water desalination

Zhu, Zhigao; Xu, Ying; Luo, Yifei; Wang, Wei; Chen, Xiaodong

2021

Zhu, Z., Xu, Y., Luo, Y., Wang, W. & Chen, X. (2021). Porous evaporators with special wettability for low-grade heat-driven water desalination. *Journal of Materials Chemistry A*, 9(2), 702-726. <https://dx.doi.org/10.1039/D0TA09193F>

<https://hdl.handle.net/10356/147860>

<https://doi.org/10.1039/D0TA09193F>

© 2021 Royal Society of Chemistry. All rights reserved. This paper was published in *Journal of Materials Chemistry A* and is made available with permission of Royal Society of Chemistry.

Downloaded on 26 Apr 2023 08:33:26 SGT

REVIEW

Porous evaporators with special wettability for low-grade heat-driven water desalination

Zhigao Zhu,^{†ab} Ying Xu,^{†ac} Yifei Luo^d, Wei Wang^{*,a}, Xiaodong Chen,^{*,d}

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

Against the backdrop of global water scarcity, porous interface distillation (PID) working on macroporous evaporation materials attracts ever-growing attention in sustainable desalination. It excels itself in the ability to treat high-salinity brines and employ low-grade heat energy from industrial sources or solar energy, as well as low installation complexity and cost. In recent years, PID evaporators with special wettability, such as omniphobicity, asymmetric superwettability, superhydrophobicity and superhydrophilicity, are especially effective in improving distillation efficiency and operation lifetime, and much progress is made in this aspect. To provide a comprehensive overview, this review puts forward a panoramic discussion on the recent progress of hydrophobic and hydrophilic porous evaporators with special wettability for low-grade heat-driven water desalination, with focuses on material and structural designs to improve distillation efficiency and long-term durability. Current challenges and future perspectives are also provided as a guide for future research towards the practical application of PID for sustainable water desalination.

1. Introduction

Freshwater production is one of the global concerns facing the shortage of water resources and escalating water pollution.^{1,2} United Nations estimate that more than 1.8 billion people (about 40% of the world's population) are experiencing absolute water scarcity.³ Additionally, this current trend will be more severe in the near future, driven by the ever-growing population and industrialization. To tackle this challenge, desalination technologies have been widely developed to extract freshwater from a range of high saline or contaminated waters, including seawater, brackish groundwater and wastewater, expecting to alleviate pressure on freshwater supply.⁴⁻⁸ Currently, Reversible electrochemical desalination technologies that based on the compensation of electric charge by ionic species are promising technologies for saline water treatment because of their cyclic efficiency and reversibility, but most of them are suffered from low-removal capacity, efficiency and rate.^{9,10} Capacitive deionization (CDI) worked on electrostatic adsorption via electrochemical double-layer capacitance theory and Faraday Redox reaction are also prosperously developed to treat industrial saline water, desalinate seawater and water-soluble radioactive wastewater, but the high concentration of total dissolved solids (TDS) and high fouling potential of industrial saline wastewater are two major challenges in CDI process, especially when desalination of high saline wastewater.^{11,12} Besides, reverse osmosis (RO) membrane technology is also regarded as the most energy-efficient desalination technology, accounting for 60% of global desalination capacity, because the liquid water does not undergo a phase transition with

low energy consumption. Traditional thermally driven technologies, such as multi-effect distillation (MED) and multi-stage flash (MSF), account for another 34%.^{3,13-15} Despite the popularity of RO technology, its energy source and high-quality electrical power is not conducive to reduce the carbon footprint of desalination systems. Additionally, RO is also challenged to desalinate high-salinity brines (excess of 80 g kg⁻¹) due to the increased osmotic pressure. Therefore, large quantities of wastewaters containing concentrated pollutants will be discharged. While for the traditional thermal techniques (e.g., MED and MSF), although high-salinity wastewater can be treated, these techniques are powered by high-grade heat energy (temperature generally higher than 70 °C). Unfortunately, the global waste heat is generally of much lower temperature, classified as low-grade heat. For instance, around 95% of waste heat from thermal power plants is below 42 °C and most naturally collected solar heat energy is also estimated to be below 50 °C.³ Therefore, it is urgently needed to develop a more sustainable desalination strategy capable of harvesting low-grade heat energy.

In recent years, an interesting class of interfacial distillation technology using evaporators with micrometer-sized pores, porous

Fig. 1. Qualitative comparison of porous interfacial distillation (PID) with established desalination technologies across a wide variety of desalination performance metrics. Technologies include reverse osmosis (RO), traditional thermal methods (multi-effect distillation and multi-stage flash). Three stars refer to excellent performance, and one star indicates poor performance.

^a State Key Laboratory of Urban Water Resource and Environment, School of Environment, Harbin Institute of Technology, Harbin 150090, P. R. China. E-mail: wangweirs@hit.edu.cn.

^b Key Laboratory of New Membrane Materials, Ministry of Industry and Information Technology, School of Environment and Biological Engineering, Nanjing University of Science and Technology, Nanjing 210094, P. R. China.

^c School of Ecology and Environment, Zhengzhou University, Zhengzhou 450001, P. R. China.

^d Nanyang Technological University, 50 Nanyang Avenue, 639798 Singapore. chenxd@ntu.edu.sg.

† These authors contributed equally to this work.

interface distillation (PID), draws particular attention for desalination.^{16–19} Steam is generated at the hotter air-water-evaporator three-phase interface and subsequently transferred through the colder air, driven by the vapor pressure gradient caused by the temperature difference. The porous structure in the evaporators provides low resistance paths for vapor diffusion or extends the air-water interface to improve vaporization efficiency. Since the temperature difference is the primary driving factor, low-grade heat can serve as an energy source to realize distillation desalination. PID excels itself in many aspects compared with conventional desalination methods (Fig. 1). PID can employ low-grade waste-heat energy to treat high-salinity brines (excess of 80 g kg⁻¹), which can not only lower the carbon footprint of the desalination process but also lead to additional environmental benefits.³ Additionally, PID can altogether reject the non-volatile solutes, including H₃BO₃ and then produce high-quality water, which is a challenge for RO.^{20–23} Besides, PID has a relatively small areal footprint than other thermal desalination technologies and is highly suitable for small-scale and emergency desalination applications, because of its highly compact configuration and easy-to-deploy feature.^{24, 25}

In fact, PID is not a new desalination technology. Its history dates back to the 1960s, when the first patent was issued in 1963 and the first journal article was published in 1967.^{26, 27} PID technologies are divided into two categories according to working principles, which are respectively based on hydrophobic evaporators and

hydrophilic evaporators (Fig. 2a). On the one hand, from the 1960s to around 2016, only hydrophobic porous membrane evaporators were applied in PID desalination processes due to the effective separation of clean vapor and polluted water. Water vapor can pass through the air-filled hydrophobic pores with low resistance, while the solutes and liquid water are rejected by waterproofing interface, thus achieving separation (Fig. 2a). Habitually, the hydrophobic PID was named membrane distillation (MD).^{28, 29} On the other hand, from around 2016, researchers also began to pay attention to hydrophilic porous evaporators. The water-infiltrated hydrophilic evaporators can extend the air-water interface, promote heat transfer and then improve vaporization efficiency (Fig. 2a).^{30, 31} Hydrophilic PID has no inherent separation ability, and thus commonly works in upward air-gap evaporation is in-situ generated by the photothermal effect of the evaporators' surface, functions in an upward evaporation configuration, thus very suitable for hydrophilic evaporators.³² Facing the energy and freshwater crises, most recently, in order to further qualify PID for sustainable desalination, energy utilization (distillation efficiency) and durable operation are becoming the two critical issues. It was revealed that individual wettable evaporators with unique structures were dramatically effective in improving distillation performance. For instance, hydrophobic evaporators with special wettability (omniphobic, asymmetrically superwetable, superhydrophobic/slippy, etc.) and low-tortuosity porous structure were explored to enhance the antiwetting, antifouling, anti-scaling and water production performance of hydrophobic

Fig. 2. (a) Schematic evaporation diagram in hydrophobic and hydrophilic evaporators. (b) The development history and trend of the PID based hydrophobic and hydrophilic porous evaporators.^{30, 31, 33, 34} Reproduced with permission from ref. 30. Copyright 2017, National Academy of Sciences. Reproduced with permission from ref. 31. Copyright 2017, Elsevier. Reproduced with permission from ref. 33. Copyright 2014, Elsevier. Reproduced with permission from ref. 34. Copyright 2020, Elsevier.

PID^{33, 35} hydrophilic evaporators with superhydrophilicity or exclusive interaction with water molecules were designed to improve energy utilization and durable performance of hydrophilic PID (Fig. 2b).^{30, 31} By far, there are several reviews on individual hydrophobic PID (MD) and hydrophilic PID in literature.^{3, 36-38} However, a comprehensive overview of PID (including both hydrophobic and hydrophilic PIDs) from the perspective of special wettability is still missing. Moreover, because of the different working modes of hydrophobic and hydrophilic PIDs, there is an urgent need to sort out the diverse design principles of special wettability and their relationships with the evaporator performance. Therefore, this review is aimed to provide such a panoramic discussion on porous evaporator design with special wettability and their performances for both hydrophobic and hydrophilic PIDs towards low-grade heat-driven sustainable water desalination. In Section 2 and 3, we will separately summarize the recent progress of the hydrophobic and hydrophilic porous evaporators with special wettability, which focuses on material designs to improve distillation efficiency and long-term durability. Moreover, recent challenges and prospects of PID are also presented in this review.

The recent ten years have witnessed the ever-growing development of PID based hydrophobic porous evaporators towards sustainable desalination. In hydrophobic PID, the hydrophobic evaporators act as the media for vapor transportation, which cannot be too thick to hinder the mass transfer. Therefore, the hydrophobic evaporators are generally manufactured into membrane-shape materials with separation function, and the corresponding processes are named membrane distillation (MD).³⁹⁻⁴¹ In the early stage, some commercial hydrophobic microfiltration membranes were first used for MD, but most of them are generally suboptimal due to their circuitous porous structure hindering vapor transfer and poor surface chemical features threatening by complicated water quality. In recent ten years, the flourishing research attentions were focused on problem-oriented design, especially on optimizing the structure of hydrophobic membranes to improve evaporation efficiency (water flux) and superwetable surface construction to overcome membrane fouling, membrane water-wetting, salt scaling and thus achieve operation durability.^{17, 20, 42} Therefore, in this section, we will review the progress of hydrophobic porous membrane evaporators design for improving evaporation efficiency and durability.

2. Hydrophobic porous evaporators

2.1. Improvement of evaporation efficiency

2.1.1. Design principle

The structure of the hydrophobic membrane is a crucial factor that affects the evaporation efficiency (water flux) of hydrophobic PID (MD), which can be described by mathematical equations. Generally, the flux of steam across an MD membrane is driven by a difference in the partial vapor pressure ($P_F - P_P$) between the feed side and the permeate side. The partial vapor pressure is induced by a different temperature across the membrane ($T_F - T_P$).^{43, 44} The water flux of MD membrane can be expressed as a function of membrane properties as Equation (1):⁴⁵⁻⁴⁸

$$J = B(P_F - P_P) \approx \frac{\varepsilon D_{wa}}{\tau d_m R_w T (1 + \frac{D_{wa}}{D_{wk}})} (P_F - P_P) \quad (1)$$

In this equation, J is the mass transfer flux, B is the permeability coefficient of a hydrophobic membrane that could be affected by the

Fig. 3. Schematic illustration of the membrane characteristics, including membrane porosity, thickness, material thermal conductivity. They together affect the membrane performances of the hydrophobic PID (MD).

membrane porosity (ε), tortuosity (τ), and the thickness (d_m). D_{wa} is the molecular diffusion coefficient of water vapor in the air, and D_{wk} is the Knudsen diffusion coefficient of water vapor in the membrane matrix; R_w is the specific gas constant of water, and T is the mean temperature of the membrane. Herein, (D_{wa}/D_{wk}) represents the diffusion coefficient ratio, which is inversely proportional to the membrane nominal pore diameter (d_p) when operated at a fixed temperature.⁴⁹ As can be seen from Equation (1), the water flux of a hydrophobic membrane can be significantly enhanced by increasing its porosity and nominal pore diameter or by reducing tortuosity and thickness of the membrane, as shown in Fig. 3.

An ideal hydrophobic membrane equipped with a low resistance to the transport of water vapor should meanwhile have high resistance to conductive heat transfer.⁵⁰ The heat and mass transfer simultaneously occur in the MD process. The resistance to heat transfer includes three sections: the hydrodynamic boundary layers at the feed side and permeate side, and the membrane resistance, which can be expressed by Equation (2).^{51, 52}

$$h_m = \frac{Q}{T_F - T_P} \approx \frac{J \Delta H(T_p)}{T_F - T_P} + \frac{\bar{k}}{d_m} = \frac{J \Delta H(T_p)}{T_F - T_P} + \frac{\varepsilon k_g + (1 - \varepsilon) k_m}{d_m} \quad (2)$$

The parameter h_m indicates the heat transfer coefficient of a hydrophobic membrane, ΔH is the vaporization enthalpy of water at T_p , and Δc_p is the difference between the isobaric specific heat capacity of liquid water and water vapor. The combined thermal conduction coefficient of the membrane is defined as the thermal conduction coefficient of the membrane ($\bar{k}$) divided by its thickness.

Meanwhile, $\bar{k}$ is co-determined by the thermal conduction of the membrane (k_m), porosity (ε), and the thermal conductivities of the gas trapped in the membrane pores (k_g): $\bar{k} = \varepsilon k_g + (1 - \varepsilon) k_m$.³ From the above presentation, increasing membrane porosity and the membrane thickness or reducing membrane thermal conductivity can significantly decrease the conductive component of the heat transfer coefficient. Overall, based on the above two equations, we can conclude that optimizing the hydrophobic membranes with high porosity, large pore size, low tortuosity, and adequate membrane thickness will play vital roles in obtaining high evaporation efficiency.

2.1.2. Mass and heat transfer management

High porosity, interconnected pore structure and low tortuosity of MD membrane would result in low resistance to water vapor transfer, thus finally leading to a large water flux.^{3, 35, 53, 54} To date, the most crucial factor determining the porous structure of a hydrophobic PID membrane should be the fabrication method.^{28, 42, 55, 56} In recent years,

it has been well-recognized that an increase in hydrophobicity (e.g., superhydrophobicity) would slow the rate of crack formation and the capillary attraction of the water into the pore, thus reducing the pore water wicking and wetting and then maintaining the separation ability of the hydrophobic membrane. Therefore, low-surface-energy hydrophobic polymers ($10 \sim 30 \times 10^{-3} \text{ mN m}^{-1}$) were frequently applied to fabricate porous hydrophobic or even superhydrophobic membranes for MD application, such as polytetrafluoroethylene (PTFE), polyethylene (PE), polypropylene (PP), polyvinylidene fluoride (PVDF), polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP), etc.^{20, 26, 55, 57-60} To process those polymers, the fabrication methods mainly include melt-blown, phase-inversion and electrospinning.

Melt-blown and phase-inversion are traditional methods with their advantages and disadvantages. PTFE, PP and PE show characterizations of high crystallization and non-polarity, which can only be processed via thermally induced phase-inversion and melt-blown methods by dissolving them in diluents under the condition at a high temperature over T_m , which is particularly wasteful of energy.⁶¹⁻⁶⁴ Besides, the pore size of conventional melt-blown membranes ranging from microns to tens of microns is difficult to satisfy the requirement of the MD membrane due to the lower separation efficiency.^{20, 65} Unlike the crystalline polymers such as PP, PE, and PTFE, the semi-crystalline polymers (e.g., PVDF) can be easily dissolved in some polar solvent such as *N,N*-dimethylformamide (DMF), *N,N*-dimethylacetamide (DMAc), *n*-methyl-2-pyrrolidone (NMP), acetone, tetrahydrofuran (THF), etc.⁶⁶⁻⁷⁰ Thus, the phase-inversion product including the flat sheet membrane and hollow fiber membrane obtained by the exchange of the solvent in the homogeneous polymer solution with the non-solvent was considered as the simplest, cheapest as well as the most energy-efficient for hydrophobic interconnected porous membrane preparation. Meanwhile, the porosity of traditional phase-inversion membranes can reach 70-80%, which can guarantee the evaporation surface area or channels for steam transmission.^{57, 71} However, the phase-inversion membranes with comparatively high tortuosity impede steam transformation and thus decrease the membrane water flux, as shown in Fig. 4a (i). It is worth noting that the water flux of the PID can be dramatically improved by decreasing the tortuosity of membrane evaporators.⁷²⁻⁷⁵ The leading cause for the difference of

membrane tortuosity lies in the differentiation of membrane pore-forming mechanism. Generally, the phase-inversion membranes obtained by immersing the polymer solution in a non-solvent bath, then the polymer rapidly precipitates out at the interface, finally forming a porous membrane with high tortuosity. While the melt-blown method could facilely fabricate low-tortuosity porous membranes with interconnected pores by accumulating fibers with a few microns to dozens of microns, but the membranes with large pore size were disadvantageous for hydrophobic PID membranes as shown in Fig. 4a (ii).

Electrospinning, as another method utilizing high voltage, is considered as one of the powerful and efficient methods to fabricate continuous fibers for constructing a non-woven structured nanofibrous membrane with high porosity, interconnected pores, narrow pore size distribution as well as low tortuosity, as shown in Fig. 4a (iii). The electrospinning nanofibrous membrane shows comparable water flux with the melt-blown fibrous membrane, which is higher than traditional phase-inversion membranes, as shown in Fig. 4b. Besides, the average pore size and pore size distribution of the membrane are critical parameters for the performance of hydrophobic PID because adequate pore size can well balance the membrane permeate flux and pore wetting resistance.⁷⁶⁻⁸⁰ It is suggested that the pore size of the hydrophobic membrane is generally in the range from 0.1 to 1 μm , and the optimal membrane size should be less than 0.5 μm , which can effectively prevent membrane wetting.^{19, 81} Owing to the unique characteristics of electrospun nanofibrous membranes with narrow pore size distribution and low tortuosity, the products fabricated by electrospinning combined with some special surface modification are generally considered as promising candidates for potential application in MD.⁸²⁻⁸⁴

The thickness of membranes also significantly impacts the efficiency of MD because thicker membranes have higher mass transfer resistance, resulting in a lower permeate flux.^{85, 86} Generally, the vapor flux initially increases with decreasing membrane thickness. However, a thinner membrane also reduces the thermal resistance resulting in a lower interface temperature difference, and the heat loss becomes dominant, which starts declining as a result of conduction losses when thickness goes below an optimal value.^{86, 87} Despite that the MD membrane has no strict requirement in mechanical strength, super-thin membranes can be easily destroyed by liquid flow. Thus, the adequate thickness of MD membranes should be strictly controlled to balance between permeate flux, thermal resistance as well as mechanical strength. Previous literature has reported that the optimal thickness of the hydrophobic MD membrane is ranging from 30-60 μm depending on the system size and MD module and configuration.³⁹ The preparation of ultrathin and self-standing hydrophobic MD membranes was comparatively tricky. As a result, the routes of the surface modification of a hydrophilic layer on one side of thick hydrophobic membrane surface or simple overlay of function layers are generally employed to balance the above trade-off.⁸⁷⁻⁹¹ The concept of dual-layer hydrophobic/hydrophilic Janus MD membranes was first introduced in the early 90's. Khayet and Matsuura et al. utilized the migration of hydrophobic macromolecules to the membrane surface to obtain flat-sheet hydrophilic-hydrophobic membranes.⁸⁸ Woo et al. fabricated a dual-layer nanofiber nonwoven membrane composed of an active hydrophobic PVDF-HFP layer with the different supporting hydrophilic layer made of either polyvinyl alcohol (PVA), nylon-6 (N6), or polyacrylonitrile (PAN).⁹² In the above configurations, the introduction of hydrophilic layers can effectively reduce the thickness of the hydrophobic vapor transfer layer, but meanwhile, bring a slight influence on the mechanical properties and thermal resistance of the whole membrane evaporators.⁸⁶ In other words, the design of the asymmetrical superwetttable MD membrane aimed at simultaneously solving the contradiction between the thickness of the active layer, heat conduction and the mechanical strength of the MD membrane.

Fig. 4. a) Membrane design for simultaneous increasing membrane porosity and decreasing tortuosity for maximal steam transformation. The insets of (i), (ii) and (iii) respectively illustrate the FE-SEM images of the phase-inversion membrane (PVDF flat-sheet membrane), melt-blown membrane (glass fiber membrane) and electrospinning membrane (polyvinylidene fluoride-co-hexafluoropropylene (PVDF-HFP) nanofibrous membranes). b) Comparison of the permeation flux of the hydrophobic porous membranes fabricated from different processes.^{16, 18, 33, 54, 55, 57, 72-75} Reproduced with permission from ref. 16. Copyright 2016, American Chemical Society. Reproduced with permission from ref. 33. Copyright 2014, American Chemical Society. Reproduced with permission from ref. 50. Copyright 2015, American Chemical Society.

In addition to controlling the pore structure, membrane porosity, and membrane thickness, decreasing the thermal conductivity of the MD membrane may become a viable direction to improve the thermal efficiency of MD technology further. However, maybe due to the similar thermal conductivity of the different organic polymer materials, it attracted little attention in reducing the thermal conductivity of a hydrophobic MD membrane. Since a ceramic material generally possesses extremely high thermal insulation, construction of ceramic membranes with excellent hydrophobic property, appropriate thickness, and porous structure may provide a promising route to overcome the thermal loss problem during MD.

2.2 Control of wetting, fouling and scaling

Hydrophobic evaporators used for PID are severely threatened by membrane wetting, fouling and scaling, which directly affect the membrane durability during the treatment of the challenging feedwaters.⁹³⁻⁹⁷ Membrane wetting, triggered by hot feedwater with low-surface-tension contaminants, could seriously destroy the membrane hydrophobicity after the long-term operation, resulting in the rapid infiltration of the feedwater into the hydrophobic membrane pores.^{18, 29} Membrane fouling is related to the hydrophobic contaminants such as oil droplets.⁹⁸⁻¹⁰¹ The hydrophobic foulants can easily accumulate onto the hydrophobic membrane surface and wick into the membrane pores owing to the underwater strong hydrophobic-hydrophobic interaction, finally preventing the transfer of vapor across the membrane.^{94, 102-104} The scaling problem is caused by the high concentration of salts in water, which gradually transforms from dissolution to crystallization state during the desalination process, resulting in the enhanced erosion of the hydrophobic membrane by accumulating/inlaying of salts on/into the membrane pores.¹⁰⁵⁻¹⁰⁷ Recently, the surface chemical modification method was well-recognized as the dominant strategy to overcome the above problems. Here, the hydrophobic membrane evaporators with special wettability are summarized, which will reveal the relationship between the special surface chemical feature and the resistance to wetting, fouling and salt scaling.

2.2.1. Design for water-wetting resistance

Regardless of water complexity, constructing superhydrophobic surfaces is an effective route to reduce the membrane wetting risk of hydrophobic membrane evaporators. To evaluate the wetting resistance of a hydrophobic evaporator, the parameter of liquid entry pressure (LEP) is introduced, which is the minimum transmembrane pressure for the salty feed water to overcome the hydrophobic surface and enter the membrane pores. An MD membrane with higher LEP can maximally prevent membrane wetting. The Young-Laplace equation is commonly used to express the LEP of a cylindrical capillary, which can be expressed as Equation (3):¹⁰⁸⁻¹¹⁰

$$LEP = \frac{-2C\gamma \cos \theta_{adv}}{d_{max}} \quad (3)$$

where γ is the surface tension of water, θ_{adv} is the advancing water contact angle of MD membrane, d_{max} is the maximal diameter of membrane pores, and C is the geometric factor depending on the membrane pore geometry. As can be observed from Equation (3), the LEP is inversely proportional to the maximal pore size of the membrane and proportional to the membrane water contact angle. Because decreasing the membranes pore size will render a reduction of the evaporation flux and efficiency, constructing superhydrophobic materials with extremely high contact angle (CA > 150°) is the frequently employed route. In general, the superhydrophobic evaporators were prepared by a combination of roughening a hydrophobic surface and subsequent modification with low surface energy materials. In recent years, inspired by natural structure (e.g., lotus leaf) with superhydrophobicity, various methods including the deposition of nanoparticles, etching, lithography, sol-gel synthesis,

Fig. 5. a) Schematic illustration of the possible membrane pore wetting against surfactants using the common superhydrophobic membrane. b) Contact angle measurement and c) the stability of membrane during MD using a hydrophobic glass fiber membrane. d) Schematic illustration of the possible resistance of membrane pore wetting mechanism using the superhydrophobic-omniphobic membrane. e) Contact angle measurement and f) the stability of membrane during MD using an omniphobic PVDF membrane.^{16, 34, 111} Reproduced with permission from ref.16. Copyright 2016, American Chemical Society. Reproduced with permission from ref.34. Copyright 2020, Elsevier. Reproduced with permission from ref.119. Copyright 2014, American Chemical Society.

template-assisted synthesis, and sputter deposition etc., have been developed to construct rough nano-surface by using inorganic nanoparticles (SiO₂, TiO₂, et al.), carbon nanotubes and metal oxides et al.^{35, 112-118} Then by combining with chemical modification, the superhydrophobic membranes with large LEP and better anti-wetting performance were obtained.

Indeed, the hydrophobic and common superhydrophobic membranes are still challenged by the polluted feedwater with organic contaminations having different surface tensions, e.g., shale gas wastewater. Although the common superhydrophobic membranes with interconnected pores show high water flux and excellent wetting resistance, the superhydrophobic membrane surfaces are susceptible to be wetted by amphiphilic organics with low-surface-tension.^{73, 74, 107, 111, 119} On the one hand, the amphiphilic surfactants can be easily adsorbed onto the superhydrophobic membrane surface via hydrophobic-hydrophobic interaction owing to the presence of hydrophobic tail moieties. On the other hand, the amphiphilic surfactants lowering the saline water surface tension γ and accumulating at the air-liquid interface result in the wetting of the superhydrophobic membrane.⁹³⁻⁹⁵ A schematic illustration of the possible membrane pore wetting mechanism of the hydrophobic porous membrane was displayed in Fig. 5a. The hierarchically rough membrane showed excellent hydrophobicity but was easily wetted by low-surface-tension contaminants, as shown in Fig. 5b. Once the low-surface-tension sodium dodecyl sulfate (SDS) wetted the hydrophobic surface, the wetted hydrophobic membrane has no resistance to the saline water that can quickly infiltrate the membrane pore, finally resulting in a sharp decrease in MD desalination efficiency (Fig. 5c). Since MD is a promising zero-emission water treatment technology for application in seawater desalination or industrial wastewater treatment, the application of MD technology to treat challenging wastewater that contains low-surface-tension contaminates is highly in demand.

To solve this problem mentioned above, much research has been devoted to developing omniphobic interconnected porous membranes for durable MD.^{16, 105, 119-121} The materials not only exhibit super waterproofing performance but also present outstanding resistance towards various liquid with diverse surface tension, hence providing enhanced robustness for MD separation. Similar to common

superhydrophobic membranes, omniphobic membranes require surface roughness and ultralow interface free energy. However, because wetting by low-surface-tension liquids is a thermodynamically favorable process for low energy surfaces, omniphobic surfaces need a re-entrant structure to develop a local kinetic barrier for the transition from the meta-stable Cassie–Baxter state to the fully wetting Wenzel state for low-surface-tension liquids. Additionally, the water-repellent agents for chemical modification often more robust than those applied for a common superhydrophobic surface. Usually, fluoroalkylsilane with long chains of fluoroalkyl groups were used to graft onto the hierarchical membrane surface to obtain the omniphobic membrane. The number of the fluorine atoms in the fluoroalkylsilane was generally 17, and also increasing the fluorine atoms can gradually enhance the membrane antiwetting performance. To date, fluorinated polyhedral oligomeric silsesquioxane, 1H,1H,2H,2H-perfluorodecyltrichlorosilane, 1H,1H,2H,2H-perfluorooctyltrichlorosilane, 1H,1H,2H,2H-perfluorodecyltriethoxysilane, 1H,1H,2H,2H-perfluoroalkyltriethoxysilane, 1H,1H,2H,2H-perfluorodecanethiol, perfluorodecanoic acid and perfluorooctanoic acid can all be employed for this application.^{122–134} Based on the special design, the omniphobic membrane can even repel most low-surface-tension liquids ranging at 10–20 mN m⁻¹, much better than a traditional superhydrophobic counterpart (above 20 mN m⁻¹), thus achieving excellent wetting resistance.

Lin et al. reported the first example of omniphobic membranes for application in the anti-wetting MD membrane.¹¹¹ A schematic illustration of the possible resistance of membrane pore wetting mechanism using the omniphobic porous membrane is displayed in Fig. 5d. The omniphobic membrane can repel all low-surface-tension contaminants owing to its extremely low surface energy of the membrane surface, thus preventing the MD membrane from being wetted. For the omniphobic membrane preparation, an electrostatic assembly method combined with surface fluorination was generally used to construct an omniphobic membrane surface. Taking glass fiber membrane as an example, first, the negatively charged borosilicate GF was treated with 3-aminopropyl-triethoxysilane (APTES) in toluene via hydrolysis to obtain a positively charged GF at the acidic condition, followed by immersing it in an aqueous suspension of silica nanoparticles (SiNPs). The negatively charged SiNPs were then assembled onto the GF fiber surface via electrostatic attraction and then strengthened by dealing with a toluene solution of silicon tetrachloride. Finally, the FAS grafted on the SiNPs via a hydrolysis reaction reacts significantly reduced the surface tension of a GF membrane after heat treatment at 130 °C for 1 h.¹⁶ The superhydrophobic-omniphobic membrane shows a hierarchical rough fiber surface owing to the aggregation of SiNPs. After being fluorinated by FAS, the hydrophilic GF membrane with hierarchical roughness turned from hydrophilic to omniphobic, which exhibits a significantly improved antiwetting property compared to the hydrophobic PTFE membrane, as shown in Fig. 5e. The omniphobic membranes with the capacity to repel all low-surface-tension contaminants, maintaining robust MD durability, as shown in Fig. 5f.

The membrane matrixes treated by strong acidic or alkali solution at high temperature to form the functional groups could destroy the membrane structure, further affecting the robustness of the MD membrane and lowering the LEP of the MD membrane. Considering the above issue, our research group developed a dual-bioinspired method to friendly assemble the hierarchically rough membrane surface. During the synthesis of the omniphobic polyimide (PI) membrane, the dopamine plays dual-functional roles as both the adhesive and the starting point to graft positively charged PEI. The membrane was first coated by the highly charged polydopamine/polyethyleneimine (PDA/PEI) layer. Zeta potential of PDA/PEI@PI membrane reveals the successful decoration of the highly positive charged PDA/PEI layer on the PI nanofiber surface.

After that, the positively charged PI fibrous membrane was then immersed in the negatively charged Ludox SM for 3 h in acetate buffer at pH=4.5 with an ionic strength of ~1 mM. The SiNPs can uniformly and compactly decorate on the PI nanofiber surface. After fluorination, the composite membrane shows omniphobicity towards various low-surface-tension liquids via contact angle measurement. Significantly, the dual-bioinspired method is a versatile method for materials roughness construction regardless of the materials composition, being organic materials, inorganic materials or metal materials.²⁹

2.2.2. Design for fouling resistance

Except for the water-wetting phenomenon of hydrophobic membranes during the PID process, the non-polar contaminants such as oils with ultralow surface energy can be easily absorbed onto the hydrophobic or omniphobic membrane surfaces to cause fouling problem via hydrophobic-hydrophobic interactions after the long-time operation.^{99, 135} Although the hierarchical micro/nanostructures of a superhydrophobic or superhydrophobic-omniphobic surface offer a kinetic barrier preventing the penetration of non-polar contaminants into membrane pores, the accumulation of these hydrophobic contaminants on the membrane surface underwater could block the membrane pores, finally reducing the membrane permeability.^{119, 136} Inspired by the features of marine creatures such as fish scales, it was demonstrated that a hydrophilic surface layer in combination with a hierarchically rough structure could maximally overcome the hydrophobic contaminant fouling due to the preferential affinity for water.^{137–142} Meanwhile, the construction of multi-scaled roughness on the membrane surface, according to the Cassie–Baxter equation, was also adapted to the underwater oleophobic surfaces since the trapped air in the pores was replaced by water.^{143, 144} However, for an MD process, the hydrophilic membrane surface for preventing membrane fouling and hydrophobic membrane surface for avoiding saline water permeation seems to be a contradictory requirement.

To solve this problem, an asymmetrically superwetttable hydrophilic/hydrophobic Janus membrane where an ultrathin hydrophilic layer was deposited onto the hydrophobic membrane surface was first proposed by Wang et al. to prevent membrane fouling.⁹⁸ Once the hydration layer formed onto the hydrophobic membrane surface, the hydrophobic contaminants were difficult to replace the infiltrated water owing to the comparable high liquid surface tension. Thus, the hydrophilic layer acts as a block wall to hinder the transport of non-polar contaminants to the membrane surface. Meanwhile, the saline water can permeate the hydrophilic layer without any barrier and vaporize at the hydrophobic membrane surface, further permeating across the hydrophobic membrane and condensing at the cold side. However, owing to the hydrophobic rather than omniphobic PVDF membrane surface, those mentioned above hydrophilic/hydrophobic Janus membrane surface with asymmetrical wettability is also proven to be wetted by low-surface-tension hydrophilic contaminants. To simultaneously solve the membrane wetting and fouling resulted from the low-surface-tension hydrophobic and hydrophilic contaminants, the omniphobic membrane surface in combination with a hydrophilic skin layer was successfully fabricated.¹³⁶ Schematic illustration of the possible simultaneous resistance of membrane pore wetting and fouling mechanism using the hydrophilic/omniphobic Janus membrane is shown in Fig. 6a. The surfactant stabilized oil-in-saline water emulsion was first demulsified at the hydrophilic skin layer of the hydrophilic/omniphobic Janus membrane surface. Then the saline water with hydrophilic contaminates permeated and evaporated at the omniphobic membrane surface, while the omniphobic membrane with the capacity to resist the low-surface-tension contaminants. The synergistic effect of the hydrophobic skin layer and the omniphobic

membrane substrate can simultaneously resist membrane wetting and fouling for MD.

Fig. 6. a) Schematic illustration of the possible simultaneous resistance of membrane wetting and fouling mechanism using the hydrophilic/omniphobic Janus membrane. Schematic illustration of a two general method to fabricate hydrophilic skin layer onto the omniphobic membrane surface, b) spraying using an air-driven spray gun and c) spraying using electrospinning. FE-SEM images of hydrophilic skin layer fabricated via spraying using d) air-driven spray gun and e) electrospinning method, respectively. (the inset in e showing the underwater oil contact angle of electrospayed hydrophilic skin layer) Comparison of the resistance of membrane wetting and fouling when treating the surfactant stabilized oil-in-saline water microemulsion of relevant membranes after MD operation, f) air-driven spray gun method and g) electrospinning method.^{34, 70, 136} Reproduced with permission from ref.34. Copyright 2020, Elsevier. Reproduced with permission from ref.74. Copyright 2018, Elsevier. Reproduced with permission from ref.136. Copyright 2017, American Chemical Society.

In order to improve the water flux, the electrospun fibrous membrane with high porosity, control pore size, and adequate thickness is recognized as the optimal candidate for application in MD.^{145–147} However, for a Janus membrane fabrication, scientists have focused on how to make a breathable and asymmetrically superwetable Janus membrane for maximal vapor transformation across the omniphobic membrane. Generally, the fabrication of Janus membranes can be done in two approaches: asymmetric decoration and asymmetric fabrication.^{148, 149} The asymmetric decoration is relatively difficult because the decoration side of the membrane should not wet the opposite wettable side during preparation, which significantly increases the difficulty of preparing an asymmetrically wettable Janus membrane. Thus, the most straightforward approach is to perfectly combine the hydrophilic layer with the omniphobic membrane without defects. In the initial stage of fabricating the asymmetrically superwetable Janus membrane, Huang et al. made utilization of spraying coating technique depositing a hydrophilic layer on a hydrophobic/omniphobic membrane surface; however, the breathability of the Janus membrane was severely sacrificed because the composite hydrophilic solution was complicated to uniformly dispersed on the hydrophobic/omniphobic membrane surface as shown in the schematic diagram of Fig. 6b.¹³⁶ More importantly, the agglomerates severely limited the transfer of vapor across the hydrophobic membrane, thus decreasing the permeation water flux. Recently, our group reported a sequential electrospinning and electrospinning technique to obtain an asymmetrically superwetable Janus nanofibrous membrane.⁷⁰ First, necklace-structured PVDF membrane substrates were fabricated via electrospinning of a LiCl/PVDF solution. Second, the Janus skin layer with asymmetric superwettability was prepared via a sequential electrospinning technique using the prepared F-SiO₂@PVDF-HFP/PS and SiO₂@PAN solutions. Owing to the strong electrostatic interaction and unevaporated solvents during electrospinning, the triple layers of the Janus membrane can be bind compactly without stratification. The PVDF nanofibrous membrane acted as membrane substrate also showed a hierarchical roughness structure via regulating the electrospinning solution. Unlike the general method using the air-driven spray gun for hydrophilic layer coating, the electrospayed

beads-on-string structured superhydrophilic skin layer on the membrane substrate cannot block the membrane pores as schematically shown in Fig. 6c, guaranteeing the vapor transformed across the omniphobic membrane without obstacles. FE-SEM images of the above two methods for preparing the hydrophilic skin layer on the omniphobic Janus membrane are also shown in Fig. 6d and e. The spraying of hydrophilic CTS-PFO/SiNPs onto the omniphobic PVDF fibrous membrane could seriously destroy the omniphobic membrane pores, thus significantly enhancing the membrane tortuosity and decreasing the water flux of the hydrophilic/omniphobic Janus membrane. Moreover, the hydrophilic layer also behaved unstable after 2 h of operation, and the curve decreased obviously after 10 h of MD operation, which was triggered by the swelling of the hydrophilic layer, as shown in Fig. 6f. Instead, the electrospaying of SiO₂@PAN skin layer onto the omniphobic substrate can maintain robust breathability of the hydrophilic/omniphobic Janus membrane and show an extremely high underwater oil contact angle of 166°, which shows robust anti-oil fouling resistance as shown in Fig. 6g.⁷⁰ The breathable and asymmetrically superwetable Janus membrane showed high water flux, good salt rejection efficiency, as well as excellent stability.

Fouling is a persistent problem where contaminants deposit on the membrane surface or inlaid in the membrane pores and could be detrimental to the membrane separation flux and efficiency.^{95, 96} In the present development, most of the hydrophobic MD reports in the literature focused on how to increase membrane flux and rejection efficiency, but very few works report an in-depth study of the fouling mechanism during the MD desalination process. In the future, understanding the fouling phenomenon and the formation mechanism of the evaporators under different water conditions should be the primary step to solve the membrane fouling problem.

2.2.3. Design for scaling resistance

If MD is applied for the high salinity wastewater treatment, membrane scaling may become a major and unavoidable technical challenge because the feed solution will eventually become oversaturated.^{150–152} The formation of mineral scales deposited on the membrane surface or inlaid in the membrane pores can both induce scaling that reduced water flux and amplify wetting that reduced salts' rejection efficiency. A schematic illustration of the possible anti-scaling behavior over the superhydrophobic membrane with a slippery surface is shown in Fig. 7a. According to the Cassie-Baxter modes, the hierarchically roughened membrane with a superhydrophobic surface reduced the contact area between the liquid and the solid contact area greatly reduced the area of the interfacial crystallization at the water-solid interface. The superhydrophobic membrane surface with minimum pinning (slippery surface) could effectively reduce the overall adhesive interaction between the crystallized layer and the membrane surface, contributing to less deposition and formation of crystal particles in the membrane pores. Instead, the superhydrophobic/hydrophobic membranes without slippery surfaces are more susceptible to membrane scaling because of their high adhesive force toward high salinity water. Recently, Xiao et al. investigated a slippery mechanism to understand the scaling behavior in membrane distillation for NaCl. They first designed a novel superhydrophobic PVDF membrane with micro-pillar arrays (MP-PVDF) using a micromolding phase separation method in combination with CF₄ plasma treatment.^{153, 154} The resultant membrane showed a high water contact angle of $175.57 \pm 1.3^\circ$ and a sliding angle of 3.0° , which displayed excellent anti-scaling in concentrating the saturated NaCl feed, as shown in Fig. 7b. Instead, both the MP-PVDF membrane without fluorination (high water contact angle, high sliding water angle) and the C-PVDF membrane (low water contact angle, high sliding water angle) can be easily scaled owing to their high adhesive toward high salinity water. It was

Fig. 7. a) Schematic illustration of the possible antiscaling and scaling mechanisms of the superhydrophobic membrane with and without slippery surfaces. b) The desalination performances of relevant membranes and c) photographs of relevant membranes after concentration of the high salinity NaCl solution.¹⁵³ Reproduced with permission from ref.153. Copyright 2019, Elsevier.

also found that the scaling of the MP-PVDF membrane was mainly occurred at the membrane surface, while for the C-PVDF membrane, the liquid was penetrated the membrane pores. Besides the phase separation method, the electrospinning and electrospraying approach also contribute to scaling-resistant superhydrophobic/slippy membrane evaporator. Lin's group fabricated an electrospun membrane with both robust anti-scaling performance and higher water flux (about 27.4 L m⁻² h⁻¹),¹⁵⁵ which was far higher than that of the phase conversion membrane (17.0 L m⁻² h⁻¹) under the similar desalination condition. Moreover, the mechanism of scaling resistance enabled by the superhydrophobic and slippy electrospun membrane was also be systematically discussed, mainly due to the roles of the surface-bound air layer in reducing the crystal-membrane contact area, nucleation propensity, and ion-membrane contact time.

The high salinity wastewater is generally complex in composition. The gypsum scaling during MD is trickier than that of NaCl. In 2018, Karanikola et al. initially found that when treating the gypsum solution, the superhydrophobic and slippy membrane surface can delay the gypsum scaling during desalination compared to the pristine PVDF membrane and the modified PVDF membranes. However, both flux decline and pore wetting had occurred, demonstrating the superhydrophobic and slippy membrane surface can only delay but not eliminate gypsum scaling.¹²⁰ Similar results were also demonstrated by Su and Xiao et al.^{154, 155} The exact mechanism underlying these different scaling behaviors between NaCl and gypsum is still unclear at this stage and needs further elucidation. To better improve the membrane gypsum scaling resistance, Horseman et al. developed a superhydrophobic and slippy membrane surface combined with a gas purging technique to effectively mitigate gypsum scaling in MD, enabling MD to concentrate a highly saline feed stream by 5-fold without suffering flux decline.¹⁵⁶ Currently, periodic purging may become the optimal and effective method in removing the deposited gypsum particles on/into the membrane surface/pores since the superhydrophobic and slippy membrane surface could effectively minimize but not eliminate the formation of crystal "anchors" within the membrane pores. A similar result was also confirmed by Ye et al.¹⁵⁷

2.3 Self-heating superhydrophobic evaporators

Besides optimizing the water flux and durability, managing the energy input by evaporator design should be another critical strategy to promote evaporation energy efficiency. In general cases, low-quality waste heat is expected to drive the MD process, leading to an energy-effective result for desalination, but waste heat sources are accessible only in some special districts in the world. So, in order to expand the application of MD with high energy efficiency, searching for proper heating routes is attracting ever-growing research attentions. In recent

years, researchers have begun to focus on the self-heating evaporators, which can in-situ generate localized low-grade heat by Joule heating effect or photothermal heating effect at the evaporator interface to drive distillation.^{158, 159} This interfacial self-heating route can reduce membrane temperature polarization or obtain higher thermal recovery compared to the bulk heating system driven by high-quality electric energy. Joule heating works by the resistance of a conductor to electron flow producing thermal energy. For Joule heated hydrophobic membrane, the membrane surface is modified by a conductive material with proper resistance (e.g., carbon nanotubes, graphene hybrid architectures, and metal nanowires).^{160, 161} When an electric current is applied across the conductive layer, the resistance to electron flow, such as ohmic heating or resistive heating can produce thermal energy for liquid water evaporation on the membrane surface. Unlike Joule heating, photothermal heating utilizes light absorbers for capturing the light of the solar spectrum and then convert solar energy to thermal energy.^{162, 163} The in-situ produced heat evaporates the liquid water on the hydrophobic membrane surface, then promotes the vaporized steam transferred through the hydrophobic membrane pores and subsequently be collected at the cold side.

2.3.1. Photothermal superhydrophobic evaporators

The investigation of MD driven by interfacial photothermal heating began in around 2015.¹⁶⁴ Generally, the reported photothermal MD processes can be classified into two types: photothermal direct contact membrane distillation (P-DCMD) and photothermal air gap membrane distillation (P-AGMD). In the P-DCMD application, some kinds of photothermal materials with robust light adsorbing properties were developed and combined with the superhydrophobic membranes, such as carbon-based materials, plasmonic materials, conjugated polymers, and organic dyes. Here, the working mechanism of the photothermal materials will not be discussed, which will be talked later in the next section on hydrophilic evaporators. We will pay our main attention to the design of the photothermal DCMD membranes. To date, there are three configurations of the photothermal DCMD membranes, e.g., blending type, coating type, and bi-layer type. (i) In the blending type, photothermal nanomaterials were dissolved or dispersed into the polymer casting solution. After the membrane formation, the photothermal components were in-situ incorporated in the membrane matrixes. For example, Curcio et al. fabricated a superhydrophobic PVDF MD membrane filled with silver nanoparticles by the blending method. Under the illumination of UV light at 366 nm of 500 W, the membrane surface temperature was 23 K greater than the bulk water, which resulted in an 11 fold increase of the transmembrane water flux, with 0.5 M NaCl being the feed water.¹⁶⁵ The blending type had the advantage of uniform distribution of the photothermal species embedded throughout the pores of the membranes but was suffering from the drawback of limited content of photothermal components. If the photothermal materials were in low content, the illumination would be shield by the polymer; and when the excessive dosage of the photothermal materials was added, the mechanical properties of the membrane matrixes will be threatened. (ii) In the coating configuration, the as-prepared MD membranes were immersed in the solution with photothermal species. Afterward, the photothermal species permeated and then attached into the membrane pores due to some interface physical or chemical interaction. For instance, Fujiwara et al. designed a series of organic dyes modified MD membranes by a coating method.¹⁶⁶⁻¹⁶⁸ The method can control the loading contents of the organic dyes but may hinder by the relatively narrow light absorbance in the entire solar spectrum of the organic molecules. Therefore, complicated dye formula was often required to enhance light utilization.¹⁶⁷ (iii) While in the bilayer configuration, a thin sheet of photothermal materials was connected to the hydrophobic membranes. For instance, Li et al. reported a

carbon black or SiO₂/Au nano-shells covered superhydrophobic MD membrane in 2017 through deposition method, which delivered an up to 33% increase in distillate flux when irradiated at 1000 W m⁻².¹⁶⁹ Later, Halas et al. together with Li demonstrated another bi-layered MD membrane structure consisting of a thin photothermal layer of carbon black impregnated polyvinyl alcohol (PVA) electrospun film and the superhydrophobic layer of PVDF membrane.³⁰ The two layers were tightly connected by the adhesive function of polydopamine (PDA). The above two designs by Li et al. had the photothermal materials only on one side of the MD membranes. The nanophotonics-enabled solar membrane distillation system was directly and entirely driven by solar energy. A pilot-scale device with an active area of $\sim 1 \times 1$ m, when tested outdoor locally at Houston, USA, was able to produce an average clean water flux of 0.5 kg m⁻² h, under less than 1 sun (700 W m⁻²) illumination. Indeed, the study on the photothermal DCMD superhydrophobic membrane is just in the beginning. Current researches are mainly focused on the concept demonstration and combination of photothermal functions with MD membranes. The evaporator design to enhance wetting control, fouling resistance, water flux, and energy efficiency are highly desired.

Photothermal membranes for air gap membrane distillation are also of research interests. For P-AGMD, the superhydrophobic porous evaporators are floated on the water surface and meanwhile kept an air gap with the cooling collectors. In this configuration, the wetting of the superhydrophobic surface is no longer fatal because it is difficult for the liquid feedwater to go through the membrane to pollute the clean water due to the gravity factor and the air gap protection. Therefore, the pore size of the evaporators for P-AGMD can be appropriately enlarged to increase the vapor flux and energy efficiency. For instance, Wang's group fabricated a self-floating hydrophobic photothermal mesh by the in-situ polymerization of polypyrrole (PPy) onto PID stainless steel (SS), followed by grafting fluoroalkylsilane via chemical vapor deposition onto the black PPy@SS surface.¹⁶⁴ PPy was rationally chosen as the photothermal polymer because of its broad-spectrum absorption, high photothermal conversion efficiency, and its facile solution-based fabrication, which permits a uniform film coating with easy control over the film thickness. The surface wettability of the membrane was controlled to conform the Wenzel's wetting behavior, which enabled the membrane to spontaneously stay at the water-air interface and allowed for sufficient contact with the water surface to precisely heat the interfacial water under the solar irradiation. For the large pore size of the photothermal mesh (larger than 5 μ m), the vapor flux was 0.92 kg m⁻² h⁻¹ with an efficiency of 58%, under the illumination of 1000 W m⁻², which was around 4 times than that without a photothermal evaporator. Besides, black TiO_{2-x} nanoparticles fabricated by Mg reduction were also attached to mesh membranes by a spin coating method. After fluorination, a hydrophobic self-floating solar evaporation mesh was obtained, which displayed an efficiency of 50.30% at 1000 W m⁻².¹⁷⁰ To sum up, in the earlier development of photothermal distillation, researchers paid their attention to hydrophobic evaporators, with inspirations from the conventional MD configuration. In the latest three years, people gradually found that heat loss of the P-AGMD was much higher than that of photothermal hydrophilic evaporators. Therefore, recent attention is shifting to hydrophilic photothermal evaporators for solar distillation, which will be detailly discussed in the next section.

2.3.2. Electric thermal hydrophobic evaporators

Except for solar-thermal conversion on the superhydrophobic evaporator surface, Joule heating utilizing the inelastic collision between electrons (the resistance of a conductor to electron flow) to produce thermal energy is another method for in-situ thermal heat production.¹⁵⁹ Carbon nanotubes, owing to its superior thermal conductivity with high current densities of 4×10^9 A cm⁻² and thermal

conductivity of 3500 W m⁻¹ K⁻¹, have opened the opportunities for efficient exploration of Joule-heating for industrial applications. To the best of our knowledge, the low anodic potentials (< 2 V_{d.c.}) applied to the CNT-based heater result in the surface charge and water splitting and the low conversion efficiency of electric to thermal. Significantly, a CNT-based heater can only serve for non-ionizable and non-corrosive environments because direct current providing the energy to the CNT surface could result in the rapid electrochemical degradation of the CNT-based heater in an ionizable medium.¹⁵⁹ Therefore, novel CNT-based heaters that can simultaneously work in the highly ionizable and corrosive conditions for self-heating thermal desalination distillation are highly desired.

Recently, Dudchenko et al. reported a porous thin-film CNT Joule heater by layer-by-layer spray coating of a uniform mixture that crosslinked the carboxylated CNT with PVA and deposited on a superhydrophobic PTFE porous membrane, finally constructing a highly conductive, porous and asymmetrically superwetable Janus membrane.¹⁶⁰ The upper-layer CNT based heater can immediately supply thermal energy when a potential across the upper layer of the composite membrane is applied. While the hydrophobic PTFE membrane takes the role of a conventional MD membrane, preventing salty water permeation and allowing the evaporated steam transport through the hydrophobic membrane pores. The high-frequency alternating current not only can maximally prevent the formation of the fully charged EDL depending on the solution ionic strength but also generate abundant thermal energy for a large amount of production of clean water. Although the energy consumption can be further reduced by optimizing the self-healing materials, the distillation process, the full-scale MD system, and the heat-recovery strategies, the energy efficiency of MD are inherently lower than conventional desalination technologies for high-salinity brines.

For graphene structured carbon like CNTs and graphene, the high-frequency alternative current could be one of the optimal methods to alleviate the degradation of the CNTs by electrooxidation and simultaneously promote enough thermal energy for high freshwater production. The high-quality energy consumption may become the fatal shortcoming toward Joule-heating using high-frequency alternative current. Joule heating of membrane surface initiated by the electric current can combine with other methods such as the above-mentioned photothermal conversion. Recently, Cui et al. developed a hybrid heating system by a rational combination of photo-electro-thermal effect on an all-graphene hybrid architecture.¹⁷¹ The solar energy can not only be absorbed fully and converted into heat but also can simultaneously be transferred into power utilizing a solar cell to further heat up the graphene skeleton frame for a much-enhanced generation of water vapor. The hybrid photo-electro-thermal system inherited the excellent characteristics of the photothermal conversion. Meanwhile, the introduction of a solar cell by converting light to electric power avoided the direct use of high-quality electrical energy. The hybrid photo-electro-thermal system combined with the graphene evaporator achieves a high-water production rate of 2.01-2.61 kg m⁻² h⁻¹ under solar illumination of 1 KW m⁻² without system optimization. This work offers a new strategy to take advantage of solar energy through the simultaneous conversion of light to heat and light to electricity to heat for maximal efficiency.

3. Hydrophilic porous evaporators

The hydrophilic or even superhydrophilic porous evaporators began to be frequently investigated and applied in PID desalination in the recent three years, and especially play important roles in photothermal distillation.^{162, 163, 172} The hydrophilic evaporators are not able to act as the liquid water separator as the hydrophobic compartments do, and therefore cannot be effectively applied in conventional MD

configuration. Nevertheless, in photothermal distillation processes, they excel themselves in the following aspects. (i) PID based on hydrophilic materials possesses a simpler configuration. Generally, hydrodynamic systems are built to drive water transportation in hydrophobic PID systems and consume some electric energy during operation. In the hydrophilic system, water transportation can be achieved by the capillarity of the hydrophilic macropores.^{173, 174} Thus, the extra hydrodynamic system can be omitted. This feature of the hydrophilic system is particularly attractive for emergency water supply in remote rural or island areas with a lack of infrastructure and electricity. (ii) Hydrophilicity is capable of enhancing the photothermal evaporation efficiency. In hydrophilic systems, water can wet the evaporator pores well and then form a thin layer of water film across the whole evaporators, subsequently adequately assuring the air-water contact area for evaporation. In contrast, water could only evaporate from the pores of the hydrophobic membranes in MD. Considering the special application of hydrophilic porous evaporators in solar photothermal PID, in this section, the current research on hydrophilic evaporators with special wettability or special interaction with water for solar photothermal PID desalination will be detailly reviewed.

3.1. Improvement of evaporation efficiency

3.1.1. Design principle

Before understanding how to design a hydrophilic evaporator with a higher evaporation efficiency, the factors impacting the vapor-generation efficiency during photothermal PIDs were first clarified. In traditional bulk heating solar distillation processes, water is generally required to be heated to a high temperature (50–90 °C) to carry on the vapor generation. This will render a quite slow response to the solar irradiation and may result in a serious heat loss due to the high specific heat capacity of water.^{173–175} Interfacial heating solar distillation based on photothermal porous evaporators has been widely accepted and employed since 2014. In this process, the temperature of water surface responses to the sunlight ultrafast and the in-situ generated heat can be mainly confined at water/air interface across the evaporators, which leaves the bulk water with a much lower temperature, and therefore greatly reduce the heat loss. **Fig. 8** displays the energy balance scheme in a typical interfacial solar PID system, with a photothermal evaporator floating at the air-water interface. In such a system, solar radiation (E_{sun}) is the only energy input which is ultimately split into and is balanced by five energy output pathways: (i) reflective solar radiation (E_{ref}), (ii) water evaporation (E_{eva}), including sensible heat and latent heat of the evaporating water, (iii) radiative heat loss from the photothermal material/water interface (E_{rad}), (iv) convective heat loss to the overlying air (E_{conv}) and (v) heat conduction loss to the underlying bulk water (E_{cond}). The progress of the photothermal hydrophilic evaporators in the past years was mainly guided by the energy diagram shown in Fig. 8 to enhance the water evaporation rate. The E_{eva} can be expressed as Equations (4) and (5):

$$E_{\text{eva}} = \frac{m h_{\text{fg}}}{A} = \alpha E_{\text{solar}} - E_{\text{conv}} - E_{\text{cond}} - E_{\text{rad}} \quad (4)$$

$$\alpha E_{\text{solar}} = E_{\text{solar}} - E_{\text{ref}} \quad (5)$$

In this equation, m is the mass transfer flux, h_{fg} is the latent heat, α is the solar absorptance, A is the surface area of the absorber facing the sun. As shown in Equations (4) and (5), the efforts should be focused on light management by increasing the solar energy adsorption and diminishing light reflection, also on heat management by minimizing wasteful heat loss ($E_{\text{conv}} + E_{\text{rad}} + E_{\text{cond}}$), especially. Additionally, in some works, E_{eva} was modulated as well to control the evaporation.

3.1.2. Light absorbance management

In solar photothermal hydrophilic PID, a suitable light absorber material is often integrated into porous scaffolds (e.g., filtration membrane, filter paper, air-laid paper, foam, metallic mesh, fibrous membrane, etc.) to construct photothermal evaporators or the material itself forms a self-supporting evaporator. Light absorber materials can effectively harvest and convert sunlight into localized heat, and then vaporize the water across the surface pores into steam.

To date, various types of light absorber materials have been explored and are capable of assembling into interconnected porous evaporators for solar distillation, for instance, plasmonic metal nanoparticles, carbon-based materials, and semiconducting materials. Metallic nanostructures, e.g., Au, Ag, Al, and Ge can produce heat through plasmon resonance and then promote the evaporation of water.^{165, 176–179} Plasmon resonance is a unique optical phenomenon occurring on metallic structures. When the frequency of incident light matches the oscillation frequency of delocalized electrons in metals, it triggers a collective excitation of the electrons and generates hot electrons. The excited hot electrons oscillate coherently with an incident electromagnetic field inside the metallic particles, resulting in heat generation by a Joule mechanism.¹⁸⁰ Carbon-based materials, including carbon black, CNTs, graphene (GO), rGO, amorphous carbon, carbonized biomass, etc. are also of the important interests for photothermal transformation because of their broad light absorption, high stability, and lightweight.^{181–186} Carbon-based materials contain no toxic metals, possess good stability under sunlight and in water due to the aromatic bonds, and can be easily made into all kinds of structures with macro-, micro- and nanoscale morphologies desirable for photothermal distillation.^{187, 188} They adsorb widely in the entire solar spectrum due to the closely spaced energy levels of the loosely-held π electrons and then release heat by relaxing the photoexcited electrons to their ground state.¹⁸⁹ Semiconductors, including inorganic (e.g., TiO_2 , Cu_2S , and Fe_3O_4) or polymeric (e.g., PPy and polydopamine) semiconductors, are another kind of effective photothermal agents for solar distillation.^{190–193} In these materials, light is absorbed to generate electron-hole pairs or photo-excitons. When the solar light has higher energy than the bandgap of the semiconductors, the above-bandgap electron-hole pairs will be created, which would then relax to the band edges and convert the extra energy into heat through a thermalization process. Photothermal semiconductors must be with a narrow bandgap, in which the electron-hole pairs could predominantly recombine to produce heat energy. This mechanism is in sharp contrast with wide-bandgap semiconductors, where most of the light energy absorbed reemits as photons after the recombination of electron-hole pairs near the bandgap edge. The relaxation dynamics of the above-bandgap

Fig. 8. Energy balance diagram of an interfacial-heating solar evaporator in open-air condition.

charges could be quite complicated, but an acoustic-phonon scattering mechanism may dominate it.¹⁹⁴ All these light absorbers have their respective advantages and disadvantages and are frequently applied for photothermal PIDs. To achieve higher light absorbance and lower reflection, two types of structural engineering methods were often explored to manage the light energy: surface microstructure design and macroscopic structure design. Light absorbance of photothermal membranes is not only related to the texture of absorbers but also highly depends on surface microstructures of the absorber layers. The first effective method to manage the light absorbance for photothermal distillation is to create highly porous structures (Fig. 9a and b). On the one hand, photothermal materials, especially inorganic and carbon-based ones, have a refractive index greater than 2, which may render a high light reflection of over 10%.¹⁹⁵ Construction of highly porous nanostructured photothermal layers has proved to be a typical effective strategy to reduce the reflected light loss.¹⁹⁶ When the sun illuminates the photothermal materials, the light can create multiple reflections inside the pores in the highly porous structures, which will result in an extremely low effective refractive index and therefore achieves a much lower light reflection and a higher absorption (Fig. 9b). On the other hand, the incident angle of light also generally impacts the light absorbance, especially for a membrane matrix with a flat surface. In some cases, highly porous surface structures, e.g., hierarchical graphene foam and plasmonic wood, can reduce the sensitivity of the photothermal materials to the sunlight incident angle and then achieve more stable light-water conversion efficiency.^{197–199} The first example of using nanosized titanium sesquioxide (Ti_2O_3) as a novel light absorber for solar-thermal conversion was reported in our previous study. A double-layer photothermal interconnected porous evaporator contained a thin layer supported on a cellulose membrane. The fairly small bandgap (~ 0.1 eV) endows Ti_2O_3 with the ability to absorb solar energy in the full spectrum range (from 250 to 2500 nm). More importantly, the light absorption ability of conventional bulk Ti_2O_3 can be improved when the material size is reduced to nanoscale due to the enhanced light scattering among the nanoparticles in the porous structure. By combining the ultra-small bandgap and the nanosized features of Ti_2O_3 nanoparticles, a nearly 100% internal and $\sim 92\%$ external solar-thermal conversion efficiency obtained in our group outperforms most of the conventional photothermal materials.²⁰⁰ Besides the above two functions, some well-designed nanoporous structures can broaden the absorption bandwidth to enhance the evaporation. Aluminum is

abundant in the earth with low cost, but the narrow response only in the UV region caused by the larger plasma frequency than Au and Ag, making Al challenged to be applied in solar distillation. Zhu et al. reported a 3D porous membrane with the attachment of self-assembled aluminum nanoparticles (NPs) in the pore arrays of the anodic aluminum membrane (AAM) by physical vapor deposition for plasmon-enhanced solar desalination (Fig. 9c). The self-assembled Al NPs tend to be closely packed along the sidewalls of the nanopores of the AAM, resulting in a strong hybridization effect of localized surface plasmons. Moreover, there is a thin layer of naturally formed aluminum oxide coated on Al NPs, which not only provided protection for stability without extra encapsulation but also significantly changed the effective dielectric of the surroundings and, therefore, further broadens the absorption bandwidth towards the IR range. In the whole solar spectrum, the effective absorption is over 96%, and the steam generation efficiency reached up to 88.4% under 4 sun illumination.²⁰¹

In addition to highly porous structures, specific surface nano-geometry could also be explored to improve light-to-heat conversion by enhancing light absorbance and reducing reflection. For example, Liu et al. developed black gold dimers with nanorods connected to nanospheres and then assembled them on a filter paper as an evaporation membrane. This evaporator exhibited high light absorbance and efficient solar evaporation because of the dimers accumulated broadband electromagnetic energy in the kissing point between the nanorod and the nanosphere, regardless of light impinging frequency, input angle, and polarization.²⁰³ Bae et al. proposed an interesting concept of self-aggregated metallic nanowire bundle array structure on the interconnected porous substrate to produce flexible thin-film black gold membranes (Fig. 9d). The multiscale structure has a sufficiently small taper angle. Hence there is a wide range of metallic nanoscale gaps from zero to hundreds of nanometers over a few microns' depths and 3 mm funnel-shaped structures. The small taper angle and varying nanogaps between aggregated metallic nanowires are responsible for the broadband absorption in the visible to near-infrared region. The 3-mm funnel structure yields ultra-broadband absorption up to 17 mm in the mid-infrared region. The thin-film black gold membrane absorbed the light with the average absorption of around 91% in the wavelength of 400 to 2,500 nm, and the solar-vapor conversion efficiency is up to 57% at an incident intensity of 20 kW m^{-2} .¹⁷⁷

Besides designing surface microstructure to improve light energy management, optimizing the surface macroscopic architecture for the evaporators sometimes is an effective alternative strategy, as illustrated in Fig. 9e. For instance, a 3D cone-shaped porous photothermal transpiration graphene oxide membrane with a 1D water path was demonstrated, which could receive light from a wide range of incident angles throughout the day.²⁰⁴ In our previous work, a porous origami as photothermal material was reported based on pencils and hydrophilic paper for solar-driven water purification.²⁰² The drawn process destroyed the Van der Waals' force between graphite layers, making pencil cartridge exfoliated into thin graphite sheets. The paper fibers combined with graphite sheets were demonstrated with broadband absorption for solar energy harvesting and excellent hydrophilicity for water transportation. The origami structure effectively promoted the utilization of solar energy attributing to the increased solar absorption area and decreased solar reflection loss compared with a planar system (Fig. 9f). Besides, the analog calculation results of COMSOL Multiphysics indicated that the origami structure was free from the dynamic solar motion. The nearly free cost, facile fabrication, and satisfactory solar-water conversion of 80% made this development promising for emergent water supply.

Fig. 9. Illustration of light adsorption & reflection over a) a flat surface. b) Illustration of light adsorption & reflection over an evaporator surface with microscopic topography, including (top inset) highly porous structure and (bottom inset) specific surface nano-geometry. c, d) are the corresponding PID evaporator examples demonstrated in the literature. e) Illustration of light adsorption & reflection over an evaporator surface with macroscopic topography and f) the corresponding evaporator example in literature.^{177, 201, 202} Reproduced with permission from ref.179. Copyright 2015, Nature Publishing Group. Reproduced with permission from ref.202. Copyright 2018, Nature Publishing Group. Reproduced with permission from ref.203. Copyright 2019, Elsevier.

Fig. 10. PID photothermal evaporators design for controlling wasteful heat loss. a) self-floating thin-film photothermal structures without a heat barrier, b) Complex structures with a water-wettable and porous support that simultaneously serves as heat barrier and unconfined water transportation route. They can be divided into bilayer structure and monolithic structure. c) Quasi-isolation multilayer structure with a water-proof heat barrier and an extra confined water channel.

3.1.3. Heat management

Controlling wasteful heat loss: As demonstrated in **Fig. 10**, besides the management of light absorbance, researchers also controlled the wasteful heat loss (E_{cond} , E_{rad} , and E_{conv}) to enhance the water evaporation of the hydrophilic evaporators. E_{rad} and E_{conv} represent radiative heat loss from the photothermal material and convective heat loss to the overlying air, respectively, the value of which are both related to the surface temperature of the evaporators and the ambient temperature. The radiative and convective losses per area are expressed by Equations (6) and (7), respectively.

$$E_{\text{rad}} = \varepsilon \sigma (T_2^4 - T_1^4) \quad (6)$$

$$E_{\text{conv}} = h (T_2 - T_1) \quad (7)$$

where ε is the emissivity, σ is the Stefan–Boltzmann constant, T_2 is the temperature at the surface of the evaporators, T_1 is the temperature of the adjacent environment, and h is the convection heat transfer coefficient. Since the steam is always surrounding the evaporator's surface during solar vaporization, the temperature difference between the two-part is generally small. Therefore, deduced from the equations, the values of E_{rad} and E_{conv} are commonly able to be suppressed by reducing the evaporators' surface temperature. Till now, the most recognized method to reduce T_2 temperature is to let competent water being transported to the photothermal evaporation surface, which can instantaneously convert the heat energy to vapor. Nevertheless, the negative result may enhance E_{cond} by decreasing heat insulation. In the past years, how to reduce the E_{cond} was attracting the most research attention. So, in this part, we will mainly discuss the control of E_{cond} , and the other two by the way.

In the early years, the bulk heating route is the main trend for solar distillation, during which photothermal dispersed agents were added into bulk water before the exposure to irradiation. This process generally presents low energy conversion efficiency of no more than 40% under unconcentrated solar irradiation.²⁰⁵ To improve energy efficiency, the solar interfacial heating concept was introduced in the early 2010s and has been a successful approach for solar water evaporation since then. At first, the photothermal materials were processed into hydrophobic ($\text{CA} \geq 140^\circ$) self-floating thin sheets or membranes, as shown in **Fig. 10a**. These interfacial heating systems deliver better energy efficiency than the dispersion configuration under the same conditions, especially under non-concentrated solar irradiation. However, the heat loss, especially the heat conduction loss (E_{cond}) from the photothermal evaporators into the underlying bulk

water driven by the large temperature difference is still a challenge. Right now, this configuration is only explored in the photothermal MD process due to the integrated feature, which has been reviewed in **Section 2.3**.

In order to further minimize E_{cond} , scientists began to develop heat localization strategy and employ thermally insulating porous heat barriers to separate the photothermal layer and the bulk water. This heat localization strategy, which restricts heat into a small amount of water in the evaporating surface, may substantially improve the efficiency of solar energy utilization. In 2014, Chen et al. proposed and designed the first bilayer structured interconnected porous evaporator by combining a graphite flake photothermal top layer on a porous carbon foam bottom layer.²⁰⁵ On the one hand, the porous carbon foam could self-float on the water surface due to the low density, and then acted as not only heat-insulating layer but also transport channels to supply water flow to the evaporation surface by leveraging capillarity. On the other hand, the photothermal exfoliated graphite layer was proceeded into hydrophilic and wettable, and it could effectively wick water to the photothermal hot spot by capillary force, thus formed a thin water films on the surface of graphite, which could result in enhanced surface area for evaporation and formation of three-phase contact lines at the edges of the capillaries. This structure yielded a high solar-to-vapor conversion efficiency of 85% that is 17% higher than that without bottom heat-insulating at 10 kW m^{-2} solar illumination and 64% at 1 kW m^{-2} by the coordination of the two parts. In 2015, Deng et al. also reported a similar work by utilizing a heat barrier of an air-laden paper to support a self-assembled Au film.¹⁷⁶ Besides heat insulation and water transportation, the bottom layer served as a substrate to improve the mechanical properties of the photothermal material, as well. After that, the bilayer structure with a porous bottom layer (**Fig. 10b**) has been widely adopted due to the improvement in energy efficiency and mechanical stability, e.g., bilayer bio-foam of rGO combining in-situ with bacterial nanocellulose, carbon nanotube membrane integrated on the silicon substrate, and so on.^{162, 206–208} The above bilayer structure could also be converted to a monolithic structure, where the heat insulation, water transportation, and photothermal in-situ heating functions were all integrated with a single layer 3D architecture. Carbon modified or plasmonic woods, plasmonic bacterial nanocellulose aerogels, rGO aerogels, rGO–polyurethane foams, carbon fibrous foams polymeric foams, and so on were prosperously reported as monolithic porous evaporators to be applied in solar PID.^{209–212} This integrated construction was claimed to be capable of decreasing the fabrication complexity and strengthening the durability for application. Nevertheless, the 3D thick porous structures with unconfined water transportation, both bilayer and monolithic architectures, are suffering from modest energy efficiency, as summarized in **Table 1**. The normal level of the evaporation efficiency is around 65% under one sun irradiation in many cases. This drawback is caused by the trade-off between heat insulation and water transportation. Water-absorbing and wettable capacity are generally required to wick water to the photothermal layer, but at the same time, the water inside the macropores greatly increases the material's thermal conduction and thus compromised the role of the thick porous structure as a heat barrier. Several impressive works can achieve a higher efficiency of 80–83% by tailing the wetting ability, porous structure, materials texture, etc. For instance, Zhu's group designed a composite aerogel comprised of rGO skeleton, carbon nanotube, and sodium alginate additives. Due to the high porosity, enhanced light adsorption (92%) by the introduction of carbon nanotubes and improved hydrophilicity by adding sodium alginate (contact angle of 74°), a quite high solar-to-vapor conversion energy efficiency of 83% at 1 sun was achieved.²¹³ Chen et al. developed a 3D nitrogen-doped porous graphene evaporator by chemical vapor deposition (CVD) with benzene or pyridine as carbon and nitrogen sources for photothermal

Table 1. The solar-to-vapor efficiency and vapor flux of various photothermal porous evaporators driven by the light energy of 1 sun. Type A, B, and C correspond to a, b, and c configuration described in Fig. 10. Type D represents the porous hydrogel evaporators that can reduce vaporization enthalpy of the water, thus increasing vapor flux.

Photothermal evaporators	Type	Year	Solar-vapor efficiency	Vapor flux (L m ⁻² h ⁻¹)	Ref
PPy mesh	A	2015	58 %	0.92	164
AAO/Au	A	2015	57 %	0.67	177
Al NP/AAM	A	2016	~58 %	~0.95	201
Graphite/carbon foam	B	2014	64 %	1.00	205
Ti ₂ O ₃ nanoparticle membrane	B	2016	\	1.32	200
Carbon nanotube wood	B	2017	65 %	0.95	208
RGO-SA-CNT aerogel	B	2017	83 %	1.67	213
Graphene oxide	B	2018	87 %	1.30	31
glass microsphere-carbon black	B	2019	82.1%	\	214
Carbon-nanotubes aerogels	B	2019	86.8%	1.37	215
PPy- straw	B	2020	81.6%	3.0	216
Carbonized mushroom	C	2017	78%	1.48	217
Graphene oxide	C	2017	87.5%	1.27	218
Carbon nanoparticles	C	2017	88 %	1.28	181
Ag/diatomite	C	2019	92.2 %	1.33	219
PDA-PEI/PPy@PI membrane	C	2019	86.9%	1.43	220
PPy cellulose paper	C	2019	90 %	1.52	221
PVA/PPy porous hydrogel	D	2019	94%	3.2	222
PVA/Graphene porous hydrogel	D	2019	95%	2.5	223
PVA-chitosan/PPy hydrogel	D	2019	92%	3.6	224
Fe-MOF/PVA hydrogel	D	2020	90%	3.2	225

steam generation via a high efficiency of 80% under 1 sun. The critical issue was that they decreased the thermal conductivity of the evaporator both by increasing the pore size via raising CVD temperature and by broadening the band gap of graphene via nitrogen doping.³² Wei and Ji demonstrated the first monolithic and wettable polymer foam composed of oligoaniline with a quite low thermal conductivity of 0.057 W m⁻¹ K⁻¹, which is times lower than that of inorganic solar absorbers. Therefore, they also obtained a high energy efficiency of 80% at 1 sun.²²⁶ However, even the above successes in decreasing in E_{cond} , the trade-off between heat insulation and water transportation cannot be eliminated, facilitating the development of another multilayer configuration with extra and confined water transportation routes. (Fig. 10c).

Zhu et al. demonstrated the first example of a multilayer configuration with confined water paths in 2016. The whole evaporator was composed of three parts, a porous and wettable photothermal sheet (GO membrane), a waterproof thermal insulator (a polystyrene foam, the thermal conductivity of ~0.04 W/mK) that simultaneously provided a floating function for the whole evaporator, and a thin layer of cellulose wrapped over the thermal insulator.²¹⁸ During the solar evaporation, efficient water supply to the photothermal spot could be enabled by the confined water path within the cellulose layer pumped by capillary force, at the same time, the

heat dissipation through water was minimized by the waterproof thermal insulator and the reduced dimensionality of the water path. The solar to vapor efficiency was kept at 80% under one-sun illumination, even with the water quantity increased, which was about 3 times that of bulk water heating system. From then on, many multilayer evaporators were designed with different kinds of confined water paths driven by capillary, for instance, 3D-printed porous hydrophilic GO pillars implanted in the body of CB/GO photothermal films (87.5% efficiency), carbonized mushroom stipes connected to the pileus (78% efficiency), filter paper strips and so on.^{31, 204, 214, 215, 217, 219, 227} Additionally, apart from waterproofing PS foam, air space was also employed as thermal insulators to promote heat localization.²²⁸ Besides, the structure and wetting feature of the top-layer photothermal materials were modulated to obtain better evaporation. For example, improving the hydrophilicity could form thin-film water evaporation near the three-phase contact line to enhance phase change. Constructing vertically aligned graphene sheets membrane with low-tortuosity channels was a benefit for releasing water vapor.^{220, 229} Now, the highest solar-to-energy conversion efficiency reported is closed to or even higher than 90%.^{181, 230-232} Our group integrated water transportation function and photo-heating function into smartly designed and low-cost photothermal membranes comprised of PPy coated hydrophilic interconnected porous paper or electrospun nanofibers.²²⁰ Afterward, these materials were coated onto a heat-insulating foam to conduct solar evaporation and achieved quite a high efficacy of over 90% under 1 sun illumination. These high values are due to the superhydrophilic feature of the evaporators, the suppressed radiative and convective losses by the good water transportation, and the minimized E_{cond} by the excellent thermal insulation. The above results are summarized and compared in Table 1.

Reducing water evaporation energy: E_{eva} is the heat energy required to achieve water evaporation, which includes two parts, sensible heat and latent heat of evaporating water. Since there are numerous hydrogen bonds between water molecules, a certain amount of E_{eva} must be required to overcome the association between water molecules to achieve phase change. Therefore, although wasteful heat energies have been effectively minimized and the solar-to-vapor efficiency has surpassed 90% under no solar concentration by optimizing the hydrophilic porous evaporators, the vapor generation fluxes are still quite low in the current reports, generally below 1.6 L m⁻² h⁻¹ under one sun illumination at room temperature. This low mass (vapor) flux is a bottleneck for the solar thermal driven PID for practical application. Equation (8) describes the relationship between vapor flux, energy efficiency, and vaporization latent heat (enthalpy) of water:

$$\eta = m\Delta H / C_{\text{opt}} P^0 \quad (8)$$

where m is the mass flux, ΔH is the vaporization enthalpy of water, P^0 is the solar irradiation power, and C_{opt} refers to the optical concentration on the absorber surface. Obviously, in an evaporation system, if η , C_{opt} , and P^0 were assumed to be constant, the vapor generation flux would be increased with decreasing water vaporization enthalpy.

Yu, Qu, and Yang demonstrated successful cases in their published works by rationally designing a porous hydrogel evaporator to reduce the value of ΔH and then dramatically boost the water generation rate.^{222, 223, 225, 233} The evaporators were constructed by self-floating porous polyvinyl alcohol (PVA) hydrogels with PPy or rGO solar absorbers incorporated into the networks. The internal gaps, micron channels as well as the hydrophilic PVA molecular meshes inside the hydrogels, could effectively facilitate the rapid replenishment of water to the evaporation surface by capillary pumping. Simultaneously, by controlling the water loaded ratio in the photothermal hydrogel, the in-situ generated heat energy was confined to only heat the water inside the evaporator. Moreover, the PVA chain could nearly eradicate the convective heat loss of water

during evaporation. Therefore, under the above heat and water management, the solar-to-vapor efficiency was improved to around 94% and 95% in the PVA/PPy and PVA/rGO hydrogel system, respectively. Indeed, the most important breakthrough of the above work is that they successfully reduced the vaporization enthalpy of water in the hydrogel molecular meshes rather than they only achieved the high solar-to-vapor energy efficiency. In the photothermal PVA hydrogel, there was a relatively high ratio of intermediate water detected by Ramon Spectra that could be attributed to the surface tension induced hydrogen bonds weakening. During evaporation, these intermediate water clusters would consume less energy than the fully bonded water cluster to achieve phase change, reducing the evaporation enthalpy (Fig. 11a). In addition, water can be evaporated as either a single molecule or in small clusters consisting of a few to tens of molecules. When water molecules were confined in the PVA molecular mesh, they were more likely to escape the polymer network as small clusters rather than individual molecules, which has been testified by tracing Li^+ that can be evaporated outside together with water clusters and collected in condensate water. As such, the water is evaporated to a state with a lower enthalpy change than conventional latent heat (Fig. 11b). Based on the above two reasons, the evaporation enthalpy of water in the evaporator was decreased by near to 50%. A high evaporation flux of $3.2 \text{ L m}^{-2} \text{ h}^{-1}$ was obtained under 1 sun solar radiation, which was almost two times higher than the previously reported record (Table 1). Furthermore, the interconnected porous evaporator exhibited a daily solar water purification yield of 20.3 L m^{-2} under natural sunlight in Florida, USA, indicating its potential for practical applications in the future. Since the above work, several other reports based on porous hydrogel evaporators with reduced vaporization enthalpy of the water were reported, which all presenting higher water flux of over $2.5 \text{ L m}^{-2} \text{ h}^{-1}$ under 1 sun solar radiation.^{224, 234} There are also some energy recycling routes to improve the efficiency of the whole systems, e.g.,

recycling the dynamic energy of water vapor by the piezoelectric effect, converting the interfacial solar steam enthalpy into electricity by thermoelectric conversion.^{135, 227, 235, 236} Because these works are focused on vapor collector design rather than PID evaporator construction, the details will not be discussed in the present manuscript.

Exploiting heat energy in other performance: Despite the great successes of reducing E_{eva} by the well-designed photothermal hydrogel evaporators, more economical and simple routes are still urgently demanded to overcome the limitation of water production effectively. Actually, in solar evaporation under real solar irradiation, there should be two parts of solar energy that can contribute to evaporation. One is the light energy, which irradiates on the confined evaporator surface and transfers to localized heat energy by the photothermal materials. This part of the energy is real-time and localized solar light energy, named as E_1 . The other is the solar heat energy independent of irradiation time and space (E_2), imported from the sun and stored as heat energy in the environment, which acts as the decisive factor to determine the environmental heat energy. As illustrated in Fig. 11c, in 2D planar evaporation (most of the current evaporators), E_1 is the main energy source to drive steam generation. While even the energy efficiency of E_1 by 2D planar evaporators has approached its limit (100% of E_1), the steam generation flux is still at a relatively low level. In this case, three-dimensional (3D) evaporators that extended a section into the atmosphere were currently proposed to enhance evaporation by capturing some E_2 .²³⁷⁻²³⁹ The evaporators could produce vapor below room temperature and then induced E_2 to flow towards the evaporation surface. Yet, the evaporation enhancement contributed by E_2 was generally less than 40%. This is reasoned that the capillary water lifting and distribution in common 3D evaporators were suppressed by the circuitous pores, which resulted in a limited energy transmission path of E_2 . Theoretically, in the same occupied space, the higher the water is transported to the 3D space by the solar evaporator, the more of the E_2 could be captured due to the extended heat transmission path, and then producing more vapor during solar evaporation.²¹⁶

In our recent work, a nature-inspired low-porous-tortuosity 3D evaporator was demonstrated to improve water production significantly. The solar evaporator, prepared from polypyrrole-modified maize straw (PMS), has upright vascular structures enabling high water lifting and horizontal micro-gaps, facilitating broad water distribution to the out-surface. Consequently, the PMS evaporator dramatically enhanced the utilization of E_2 for promoting evaporation. The maximum vapor generation rate of a single PMS ($10.3 \text{ L m}^{-2} \text{ h}^{-1}$) respectively increased 2.5 times and 6 times compared with the conventional 3D evaporators and the planar evaporators of the identical occupied area. A scaled-up PMS array achieved a state-of-the-art vapor generation rate of $3.0 \text{ L m}^{-2} \text{ h}^{-1}$ under the simulated condition and record-high clean water production of $2.2 \text{ L m}^{-2} \text{ h}^{-1}$ for actual seawater desalination under natural conditions (one-sun intensity). Moreover, due to the robust water transmission channel in superhydrophilic PMS, there was no salt scaling formed during long-term desalination. This breakthrough reveals great potentials for cost-effective freshwater production and the rational design of high-performance photothermal evaporators for solar distillation.²¹⁶ Beside solar heat energy, the additional solar electricity is also a useful energy form for evaporation. A photo-electro-thermal evaporation system based on solar evaporation system and solar cell, which reached a record high water-generation rate of $4.73 \text{ kg m}^{-2} \text{ h}^{-1}$ under 1 sun illumination, was reported in our recent study.²⁴⁰

3.2. Control of salt scaling and organic pollutants

Despite the improvement of the evaporation efficiency, several new challenges associated with practical applications of solar photothermal PID need to be addressed. As a kind of raw water widely

Fig. 11. The water evaporation diagrams across common photothermal evaporators a) and hydrogel photothermal evaporators with hydrogen bonds weakening function b). Less energy was consumed in b) case than a) case to achieve the same water generation due to intermediate water clusters in the liquid water phase and small water clusters in the vapor phase. c) The schematic illustration of the solar energy utilization among typical planar system, a conventional 3D cylinder with circuitous pores and PMS with low-tortuosity in water evaporation.²¹⁶ Reproduced with permission from ref.218. Copyright 2020, American Chemical Society.

used in solar-driven PID, seawater contains 3.5 wt% total dissolved solids, including NaCl, CaCl₂, and MgCl₂, which would probably precipitate into crystals when water quickly evaporates from the surface of the solar evaporator. These salt crystals build-up would clog the porous structures and accumulate during continuous operation, resulting in the reduction of evaporation rate.²²³ Especially, the hydrophilic photothermal PID systems with higher evaporation flux are suffering from stronger interaction with inorganic salts than the hydrophobic counterparts.²⁴¹ Therefore, salt crystallization as a kind of structure fouling problem in interconnected porous structures needs to be solved. To overcome this issue, many investigations have been carried out for the development of salt-resistant photothermal materials. These strategies have been proven to be very efficient to this end, which may mainly follow three kinds of salt elimination mechanisms.²⁴²

The first one is the ex-situ produce of the salt crystals, relying on the law of salt migration (Fig. 12a and b). According to the research of Mi et al., who reported a relatively smooth hydrophilic graphene leaf as the solar evaporator. The salt crystallization would block the surface of the evaporator and reduce the steam production if without any particular structure design.²²⁸ Hence, the ex-situ produce of the salt crystal is an attractive solution.²⁴³ One way to adjust the salt position is depending on the changes in water conservancy conditions. Zhang et al. designed an evaporator composed of a horizontal evaporation disc and a vertical solution uptake thread. The crystallization sites of the salt on the disc were spatially isolated from the water evaporation surface, achieving continuous steam generation and salt harvesting in over 600 hours of non-stop operation.²⁴⁴ Another way is separating the evaporation surface and crystallization surface. Shi et al. designed a 3D cup that produces vapor in the cup inner-wall and solid salt as a by-product at the cup outer-wall. By rationally separating the light-absorbing surface from the salt precipitation surface, the 3D cup can be operated at high salt concentration without noticeable water evaporation rate decay in long-term operation along with continuous salt crystals removing from the cup wall.²⁴⁵ Although the above methods required mechanical removal, it prevented reducing the evaporation rate caused by the salt concentration. The formed salt crystals were recovered in solid-state, thus can achieve the goal of zero liquid discharge.

The second strategy is in-situ melting of the salt crystals, relying on the salinity gradient (Fig. 12c). Differing from the principle that hydrophobic interface prevents the formation of salt scale (separating the evaporation interface from the cold saline water), the salt-rejected superhydrophilic porous structure is generally designed as an integrated structure to ensure the smooth transport of salt ions.²⁴⁶ In our previous report that named it a salt auto-cleaning solar evaporation, and the principle and structure-activity relationship of the salt auto-cleaning process was revealed in detail. Thermodynamically, the movement of crystallized salt back to bulk seawater is a spontaneous process driven by the potential chemical difference of different salt concentrations. The intrinsic properties of its structure also determine the kinetic factor of the salt diffusion in the membrane. According to Fick's Law, if the salt auto-cleaning process was considered as the ions diffusing in the porous medium, the structure of the auto-cleaning material must possess a high porosity and superhydrophilic feature with large water content to increase the diffusion coefficient of salt ions to migrate back to the bulk water.²²¹ Besides, other groups reported similar results.^{223, 246-248} For instance, Chen et al. demonstrated an integrated evaporator with hydrophilic black cellulose fabrics as a light absorber, hydrophilic, while fabrics as water transporter and expanded polystyrene foams as an insulator.²⁴⁸ In this evaporator structure, the hydrophilic feature offered abundant water channels for rapid salt ions exchanging, leading to the rejection of salt scaling (Fig. 12d). Zhu et al.

demonstrate a water lily-inspired hierarchical structure that enables efficient evaporation (~80% solar-

Fig. 12. a) The schematic diagram of the salt scaling formed on the photothermal evaporators. b) Mechanical removal of the formed salt crystals. c) Cleaning the formed crystal automatically relying on the salinity gradient, and d) the corresponding multilayer porous evaporators with abundant salt and water migration channels. e) The formation of concentration difference during evaporation and f) the diagram of electricity generation driven by salinity gradient.^{228, 245, 249, 250} Reproduced with permission from ref.230. Copyright 2017, American Chemical Society. Reproduced with permission from ref.247. Copyright 2018, American Chemical Society. Reproduced with permission from ref.249. Copyright 2017, Royal Chemical Society. Reproduced with permission from ref.250. Copyright 2018, Royal Chemical Society.

to-vapor efficiency) out of high-salinity brine and wastewater containing heavy metal ions (30 wt %). The upper epidermis absorbs sunlight and provides stomata for water vapor escape, and has the well-known hydrophobic self-cleaning property. The self-floating leaf exists a confined water path provided by vascular bundles to pump up water and then spread it into the large surface of the water lily.²⁴² Besides, taking advantage of both hydrophilic and hydrophobic porous evaporators and their structure optimization, researchers have developed salt-resistant solar evaporators with multifunctional layers. Zhu et al. reported a flexible and salt-resistant Janus absorber with the upper hydrophobic carbon black nanoparticles coating polymethylmethacrylate layer for light absorption and bottom hydrophilic polyacrylonitrile layer for pumping water. Salt may only be deposited in the hydrophilic PAN layer and quickly be dissolved back automatically, attributing to the salinity gradient between saturated brine and bottom seawater.²⁵¹ Similarly, Yang et al. reported a hydrophobic salt-resistant hierarchical layer of Cu₂SnSe₃ (or Cu₂ZnSnSe₄) nanosphere arrays for broad solar harvesting and a hydrophilic filter membrane for continuous water supply and vapor generation. The as-fabricated devices achieve remarkable and stable solar steam efficiency of 86.6% under one sun.²⁵²

The third strategy to tackle the salt scaling problem is energy and resource recovery. Rather than being regarded as a pollutant, the salt in the solution can also be utilized as an alternative source of energy. Through inducing a high salinity gradient between the concentrated seawater and raw seawater, Yang et al. reported a new kind of technology to generate extra electricity while evaporating water (Fig. 12e and f). By employing a hydroid system of carbon nanotube modified filter paper and a commercial Nafion membrane, the system achieved a solar thermal efficiency of 75 % and extra electricity power of 1 W m⁻² under one sun illumination.²⁴⁹ Although electricity production is at a low level, the design still provides a new idea for blue energy utilization in PID. Besides the above methods, the

crystallization sites of salt could also be spatially isolated from the water evaporation surface by the hydraulic scour of microfluidics in confined water paths, thus achieving continuous steam generation and salt harvesting in over 600 hours of non-stop operation.²⁴⁴

In addition to the common salt problem, raw water for solar evaporation also contains organic matters, which would deteriorate the water quality and form fouling materials on the evaporator surface.²⁵³ Rather than relying on extra contamination removal process, Lou et al. firstly prepared a porous-structure based on rGO and TiO₂ composites allowing for facile incorporation of photocatalysts to regenerate clean water out of contaminated water and solar-driven PID.²¹² Subsequently, the black Ti₂O₃ and TiO_x designed by Wang et al. also possess the function of photocatalysis as well as photothermal evaporation, although the mechanism was not detailly investigated in their papers.^{170, 200} In fact, with reasonable water transportation and heat preservation structure design, the interconnected porous evaporator that attached various photocatalysts would all possess the property of degrading organic matter in raw water.²⁵⁴⁻²⁶⁰ It should be pointed out that the abovementioned works only reported the phenomena of photocatalysis in the solar-driven PID, and the currently targeted contaminants are generally nonvolatile model organic matters, such as dyes. While these works may be meaningful for evaporator fouling control (though not discussed in the articles), it is still unknown whether photocatalysis is effective for volatile organic compounds rejection, which is important to the cleanness of condensate water.

4. Conclusions

In summary, this article reviewed the recent development of evaporators with special wettability for porous interface distillation in the past ten years. For PID processes working on hydrophobic porous evaporators, researchers modulated the physical structure, e.g., thickness, porosity, pore tortuosity, and size of the membranes, to steadily promote the distillation flux. At the same time, they endowed the membranes with superhydrophobic, omniphobic or asymmetric wettable surface features to enhance the running durability. In addition, the self-heating evaporators with outstanding capture ability toward in-situ generated heat were also attracting interests. On the other hand, studies on PID processes working on hydrophilic porous evaporators just boomed in the past 5 years and mainly focused on solar-driven photothermal distillation. Amazing achievements have been realized in improving solar-to-vapor efficiency to over 90% and vapor flux to higher than 2 L m⁻² h⁻¹ under one sun intensity by rationally modulating light absorbance, managing heat loss and controlling feedwater supply during evaporation in the superhydrophilic evaporators. Besides, salt scaling control and material resource recovery also witnessed noticeable progress through rational evaporator design. Despite the impressive advancement in PID in the past years, there are still many significant barriers standing before its practical application. The main challenges and opportunities are listed as follows, which call for much more research attention.

(1) Increasing water generation

Hydrophobic PID: Water flux and energy efficiency of hydrophobic porous evaporators for MD should be further improved. Specifically, driven by a temperature difference of 40 K, the water flux of MD processes is generally below 40 L m⁻² h⁻¹, which requires substantial improvement. To tackle this problem, thermal insulation materials, such as flexible ceramics, with low thermal conductivity may be processed into hydrophobic evaporation membranes, which can reduce the temperature polarization across the membrane and then enhance evaporation efficiency. Besides, membrane materials with straight porous architecture are anticipated to facilitate the mass transfer during distillation.

Hydrophilic PID: Photothermal superhydrophilic evaporators harvesting solar energy are approaching the energy utilization limit under no solar concentration, but they still suffer from modest vapor production rates (generally below 2 L m⁻² h⁻¹). Boosting the utilization of the unconfined solar heat energy or reducing the latent heat of water by evaporator design might be promising routes to solve the problem.

(2) Reducing water-wetting of hydrophobic evaporators

The antiwetting ability of the hydrophobic evaporators should be further improved towards long-term practical application. In most reports on hydrophobic PID evaporators, researchers assessed the stability of the membranes only in several hours to tens of hours. Those evaporators reported in the literature are challenged to satisfy the applicative demand in engineering due to the complex source water quality in real life. Additionally, there is a trade-off between water flux and anti-wetting performance (separation efficiency) in traditional membranes. Facing the above problems, straight porous membrane evaporators with homogeneous and superomniphobic surfaces are likely to break the trade-off, enabling fast steam transportation while minimizing aggression from hot steam and surfactants.

(3) Curbing salt scaling

More attention should be paid to salt scaling in membrane pores. The source waters for distillation in realistic scenarios, such as seawater, industrial wastewater, and hypersaline waters, contain many dissolved salt species, e.g., NaCl, CaSO₄, and MgSO₄. Their scaling behaviors, especially of Ca²⁺ and Mg²⁺, would have a heavy toll on evaporation durability but have not been detailly investigated. In the future, a deeper understanding of salt scaling on evaporators with different surface characteristics should be explored. Besides, a suitable evaporator design is to be devised to achieve a much better anti-scaling function.

(4) Guarding against volatile organic compounds

PID can completely reject non-volatile solutes at the vapor-liquid conversion interface and then produce high-quality water, thus holding great promise for clean water production, wastewater treatment, and seawater desalination. However, many source drinks of water are polluted with volatile organic compounds (VOCs). VOCs can evaporate from the interconnected pores of evaporators and be collected together with distilled water; moreover, they may even be enriched in the collected distillate during interfacial distillation, which will seriously threaten the water quality. More advanced and multifunctional PID evaporators combined with catalytic degradation function or selective permeation will alleviate this issue.

In conclusion, PID excels itself in various aspects, such as the ability to employ low-grade waste-heat energy from industrial processes, capturing solar energy at the localized interface for in-situ driven distillation, relatively small areal footprint, low-pressure operation, outstanding water quality, highly compact configuration, and easy-to-deploy feature. It is regarded to be highly suitable for small-scale and on-site desalination applications towards small sized water plants, off-grid, and point-of-use potable clean water production apparatus. By resolving the abovementioned challenges and bottlenecks, PID will be endowed with a more promising prospect for practical applications.

Conflicts of interest

The authors declare that there are no conflicts to declare.

Acknowledgements

The authors gratefully acknowledge the National Natural Science Foundation of China (Grant Nos. 52000105, 52000161, 51873047 and 51761145031), the 67th batch of China

Postdoctoral Science Foundation (No. 2020M671503), the Scientific Research Foundation of Heilongjiang Prov. (No. YQ2020B003), Singapore National Research Foundation (NRF2017NRFNSFC001-048) and CREATE Program of Nanomaterials for Energy and Water Management for the funding support.

Notes and references

1. M. Kumm, J. H. Guillaume, H. de Moel, S. Eisner, M. Florke, M. Porkka, S. Siebert, T. I. Veldkamp and P. J. Ward, *Sci. Rep.*, 2016, **6**, 38495.
2. J. Leijon and C. Boström, *Desalination*, 2018, **435**, 161-171.
3. A. Deshmukh, C. Boo, V. Karanikola, S. Lin, A. P. Straub, T. Tong, D. M. Warsinger and M. Elimelech, *Energy Environ. Sci.*, 2018, **11**, 1177-1196.
4. M. Elimelech and W. A. Phillip, *Science*, 2011, **333**, 712-717.
5. M. A. Shannon, P. W. Bohn, M. Elimelech, J. G. Georgiadis, B. J. Mariñas and A. M. Mayes, *Nature*, 2008, **452**, 301-310.
6. B. Kim, R. Kwak, H. J. Kwon, V. S. Pham, M. Kim, B. Al-Anzi, G. Lim and J. Han, *Sci. Rep.*, 2016, **6**, 31850.
7. K. C., *Environ. Sci. Technol.*, 2006, **46**, 639.
8. D. Qu, J. Wang, L. Wang, D. Hou, Z. Luan and B. Wang, *Sep. Purif. Technol.*, 2009, **67**, 21-25.
9. P. Srimuk, X. Su, J. Yoon, D. Aurbach and V. Presser, *Nat. Rev. Mater.*, 2020, **5**, 517-538.
10. V. Pothanamkandathil, J. Fortunato and C. A. Gorski, *Environ. Sci. Technol.*, 2020, **54**, 3653-3662.
11. S. Huo, Y. Zhao, M. Zong, B. Liang, X. Zhang, I. U. Khan, X. Song and K. Li, *J. Mater. Chem. A*, 2020, **8**, 2505-2517.
12. E. Liu, L. Y. Lee, S. L. Ong and H. Y. Ng, *Water Res.*, 2020, **183**, 116059.
13. N. Ghaffour, T. M. Missimer and G. L. Amy, *Desalination*, 2013, **309**, 197-207.
14. L. F. Greenlee, D. F. Lawler, B. D. Freeman, B. Marrot and P. Moulin, *Water Res.*, 2009, **43**, 2317-2348.
15. R. Zhang, J. Tian, S. Gao and B. Van der Bruggen, *J. Mater. Chem. A*, 2020, **8**, 8831-8847.
16. C. Boo, J. Lee and M. Elimelech, *Environ. Sci. Technol.*, 2016, **50**, 8112-8119.
17. A. K. An, J. Guo, E.-J. Lee, S. Jeong, Y. Zhao, Z. Wang and T. Leiknes, *J. Membr. Sci.*, 2017, **525**, 57-67.
18. L. D. Tijting, Y. C. Woo, W. Shim, T. He, J.-S. Choi, S.-H. Kim and H. K. Shon, *J. Membr. Sci.*, 2016, **502**, 158-170.
19. A. Alkhudhiri, N. Darwish and N. Hilal, *Desalination*, 2012, **287**, 2-18.
20. P. Wang and T.-S. Chung, *J. Membr. Sci.*, 2015, **474**, 39-56.
21. H. Attia, S. Alexander, C. J. Wright and N. Hilal, *Desalination*, 2017, **420**, 318-329.
22. M. T. M. Gryta and A. W. M. J. Grzechulska, *J. Membr. Sci.*, 2001, **181**, 279-287.
23. C. Li, X. Li, X. Du, T. Tong, T. Y. Cath and J. Lee, *ACS Appl. Mater. Interfaces*, 2019, **11**, 18456-18465.
24. A. Chafidz, S. Al-Zahrani, M. N. Al-Otaibi, C. F. Hoong, T. F. Lai and M. Prabu, *Desalination*, 2014, **345**, 36-49.
25. J. Woods and J. Pellegrino, *Int. J. Heat Mass Transfer*, 2013, **59**, 46-57.
26. M. S. El-Bourawi, Z. Ding, R. Ma and M. Khayet, *J. Membr. Sci.*, 2006, **285**, 4-29.
27. M. Khayet and J. I. Mengual, *Int. J. Nucl. Desalin.*, 2005, **1**, 435-449.
28. L. Eykens, K. De Sitter, C. Dotremont, L. Pinoy and B. Van der Bruggen, *Sep. Purif. Technol.*, 2017, **182**, 36-51.
29. Z. Zhu, Y. Liu, H. Hou, W. Shi, F. Qu, F. Cui and W. Wang, *Environ. Sci. Technol.*, 2018, **52**, 3027-3036.
30. P. D. Dongare, A. Alabastri, S. Pedersen, K. R. Zodrow, N. J. Hogan, O. Neumann, J. Wu, T. Wang, A. Deshmukh, M. Elimelech, Q. Li, P. Nordlander and N. J. Halas, *Proc. Natl. Acad. Sci. U. S. A.*, 2017, **114**, 6936-6941.
31. Y. Li, T. Gao, Z. Yang, C. Chen, Y. Kuang, J. Song, C. Jia, E. M. Hitz, B. Yang and L. Hu, *Nano Energy*, 2017, **41**, 201-209.
32. Y. Ito, Y. Tanabe, J. Han, T. Fujita, K. Tanigaki and M. Chen, *Adv. Mater.*, 2015, **27**, 4302-4307.
33. C. Yang, X. Li, J. Gilron, D. Kong, Y. Yin, Y. Oren, C. Linder and T. He, *J. Membr. Sci.*, 2014, **456**, 155-161.
34. Z. Zhu, L. Zhong, X. Chen, W. Zheng, J. Zuo, G. Zeng and W. Wang, *J. Membr. Sci.*, 2020, **615**, 118499.
35. X. Li, M. Wang, C. Wang, C. Cheng and X. Wang, *ACS Appl. Mater. Interfaces*, 2014, **6**, 15272-15282.
36. H. Saleem, L. Trabzon, A. Kilic and S. J. Zaidi, *Desalination*, 2020, **478**, 114178.
37. M. Gao, L. Zhu, C. K. Peh and G. W. Ho, *Energy Environ. Sci.*, 2019, **12**, 841-864.
38. C. Chen, Y. Kuang and L. Hu, *Joule*, 2019, **3**, 683-718.
39. L. Martínez and J. M. Rodríguez-Maroto, *J. Membr. Sci.*, 2008, **312**, 143-156.
40. L. Eykens, I. Hitsov, K. De Sitter, C. Dotremont, L. Pinoy, I. Nopens and B. Van der Bruggen, *J. Membr. Sci.*, 2016, **498**, 353-364.
41. H. Y. Wu, R. Wang and R. W. Field, *J. Membr. Sci.*, 2014, **470**, 257-265.
42. L. D. Tijting, J.-S. Choi, S. Lee, S.-H. Kim and H. K. Shon, *J. Membr. Sci.*, 2014, **453**, 435-462.
43. K. W. Lawson and D. R. Lloyd, *J. Membr. Sci.*, 2015, **120**, 123-133.
44. M. Khayet, *Adv. Colloid Interface Sci.*, 2011, **164**, 56-88.
45. R. Krishna, *Chem. Soc. Rev.*, 2015, **44**, 2812-2836.
46. P. J. A. M. Kerkhof and M. A. M. Geboers, *Chem. Eng. Sci.*, 2005, **60**, 3129-3167.
47. J. Lee, T. Laoui and R. Karnik, *Nat. Nanotechnol.*, 2014, **9**, 317-323.
48. A. P. Straub, N. Y. Yip, S. Lin, J. Lee and M. Elimelech, *Nat. Energy*, 2016, **1**, 16090.
49. S. K. Bhatia, M. R. Bonilla and D. Nicholson, *Phys. Chem. Chem. Phys.*, 2011, **13**, 15350-15383.
50. A. Deshmukh and M. Elimelech, *J. Membr. Sci.*, 2017, **539**, 458-474.
51. S. Lin, N. Y. Yip and M. Elimelech, *J. Membr. Sci.*, 2014, **453**, 498-515.
52. M. Gryta and M. Tomaszewska, *J. Membr. Sci.*, 1998, **144**, 211-222.
53. X. Li, C. Wang, Y. Yang, X. Wang, M. Zhu and B. S. Hsiao, *ACS Appl. Mater. Interfaces*, 2014, **6**, 2423-2430.
54. X. Li, X. Yu, C. Cheng, L. Deng, M. Wang and X. Wang, *ACS Appl. Mater. Interfaces*, 2015, **7**, 21919-21930.
55. M. I. Siyal, A. A. Khan, C.-K. Lee and J.-O. Kim, *Sep. Purif. Technol.*, 2018, **205**, 284-292.
56. R. Thomas, E. Guillen-Burrieza and H. A. Arafat, *J. Membr. Sci.*, 2014, **452**, 470-480.
57. K. Yu Wang, T.-S. Chung and M. Gryta, *Chem. Eng. Sci.*, 2008, **63**, 2587-2594.
58. M. Gryta, *J. Membr. Sci.*, 2007, **287**, 67-78.
59. H. J. Hwang, K. He, S. Gray, J. Zhang and I. S. Moon, *J. Membr. Sci.*, 2011, **371**, 90-98.
60. B. S. Lalia, E. Guillen-Burrieza, H. A. Arafat and R. Hashaiekh, *J.*

- Membr. Sci.*, 2013, **428**, 104-115.
61. B. S. Sprague, *J. Macromol. Sci., Part B: Phys.*, 2006, **8**, 157-187.
 62. D. L. Green, L. McAmish and A. V. McCormick, *J. Membr. Sci.*, 2006, **279**, 100-110.
 63. L. Huang, P. Hsu, C. Kuo, S. Chen and J. Lai, *Desalination*, 2008, **233**, 64-72.
 64. S. H. Tabatabaei, P. J. Carreau and A. Ajji, *J. Membr. Sci.*, 2009, **345**, 148-159.
 65. C. Cabassud and D. Wirth, *Desalination*, 2003, **157**, 307-314.
 66. G. Kang and Y. Cao, *J. Membr. Sci.*, 2014, **463**, 145-165.
 67. S. Rajabzadeh, T. Maruyama, T. Sotani and H. Matsuyama, *Sep. Purif. Technol.*, 2008, **63**, 415-423.
 68. P. Sukitpaneinit and T.-S. Chung, *J. Membr. Sci.*, 2009, **340**, 192-205.
 69. Y. Zhai, N. Wang, X. Mao, Y. Si, J. Yu, S. S. Al-Deyab, M. El-Newehy and B. Ding, *J. Mater. Chem. A*, 2014, **2**, 14511-14518.
 70. Z. Zhu, Z. Liu, L. Zhong, C. Song, W. Shi, F. Cui and W. Wang, *J. Membr. Sci.*, 2018, **563**, 602-609.
 71. H. Fan and Y. Peng, *Chem. Eng. Sci.*, 2012, **79**, 94-102.
 72. A. Razmjou, E. Arifin, G. Dong, J. Mansouri and V. Chen, *J. Membr. Sci.*, 2012, **415-416**, 850-863.
 73. L. Chen, A. Huang, Y. Chen, C. Chen, C. Hsu, F. Tsai and K. Tung, *Desalination*, 2018, **428**, 255-263.
 74. L. Deng, H. Ye, X. Li, P. Li, J. Zhang, X. Wang, M. Zhu and B. S. Hsiao, *Sep. Purif. Technol.*, 2018, **206**, 14-25.
 75. Y. Liao, R. Wang and A. G. Fane, *Environ. Sci. Technol.*, 2014, **48**, 6335-6341.
 76. J. Phattaranawika, R. Jiratananon and A. G. Fane, *J. Membr. Sci.*, 2003, **215**, 75-85.
 77. S. Adnan, M. Hoang, H. Wang and Z. Xie, *Desalination*, 2012, **284**, 297-308.
 78. A. Imdakm and T. Matsuura, *Procedia Eng.*, 2012, **44**, 1874.
 79. L. Martínez, F. J. Florido-Díaz, A. Hernández and P. Prádanos, *Sep. Purif. Technol.*, 2003, **33**, 45-55.
 80. L. Zhong, Z. Zhu, Y. Han, Q. Wang, D. Liu, F. Cui, B. Li and W. Wang, *Environ. Sci.: Nano*, 2019, **6**, 2553-2564.
 81. K. W. Lawson and D. R. Lloyd, *J. Membr. Sci.*, 1997, **124**, 1-25.
 82. S. H. Park, J. H. Kim, S. J. Moon, E. Drioli and Y. M. Lee, *J. Membr. Sci.*, 2018, **550**, 545-553.
 83. W. Qing, X. Shi, W. Zhang, J. Wang, Y. Wu, P. Wang and C. Y. Tang, *J. Membr. Sci.*, 2018, **564**, 465-472.
 84. Z. Dong, X. Ma, Z. Xu, W. You and F. Li, *Desalination*, 2014, **347**, 175-183.
 85. F. Laganà, G. Barbieri and E. Drioli, *J. Membr. Sci.*, 2000, **166**, 1-11.
 86. X. An, G. Xu, B. Xie and Y. Hu, *J. Mater. Chem. A*, 2019, **7**, 2376-2384.
 87. M. Khayet, M. C. García-Payo, L. García-Fernández and J. Contreras-Martínez, *Desalination*, 2018, **426**, 174-184.
 88. M. Khayeta and T. Matsuura, *Desalination*, 2003, **158**, 51-56.
 89. L. D. Tijing, Y. C. Woo, M. A. H. Johir, J.-S. Choi and H. K. Shon, *Chem. Eng. J.*, 2014, **256**, 155-159.
 90. M. Qtaishat, M. Khayet and T. Matsuura, *J. Membr. Sci.*, 2009, **329**, 193-200.
 91. N. G. P. Chew, Y. Zhang, K. Goh, J. S. Ho, R. Xu and R. Wang, *ACS Appl. Mater. Interfaces*, 2019, **11**, 25524-25534.
 92. Y. C. Woo, L. D. Tijing, M. J. Park, M. Yao, J.-S. Choi, S. Lee, S.-H. Kim, K.-J. An and H. K. Shon, *Desalination*, 2017, **403**, 187-198.
 93. M. Rezaei, D. M. Warsinger, V. J. Lienhard, M. C. Duke, T. Matsuura and W. M. Samhaber, *Water Res.*, 2018, **139**, 329-352.
 94. Z. Wang and S. Lin, *Water Res.*, 2017, **112**, 38-47.
 95. A. K. An, J. Guo, S. Jeong, E. J. Lee, S. A. A. Tabatabai and T. Leiknes, *Water Res.*, 2016, **103**, 362-371.
 96. L. D. Tijing, Y. C. Woo, J.-S. Choi, S. Lee, S.-H. Kim and H. K. Shon, *J. Membr. Sci.*, 2015, **475**, 215-244.
 97. M. Rezaei, D. M. Warsinger, J. H. Lienhard V and W. M. Samhaber, *J. Membr. Sci.*, 2017, **530**, 42-52.
 98. Z. Wang, D. Hou and S. Lin, *Environ. Sci. Technol.*, 2016, **50**, 3866-3874.
 99. G. Zuo and R. Wang, *J. Membr. Sci.*, 2013, **447**, 26-35.
 100. D. M. Warsinger, J. Swaminathan, E. Guillen-Burrieza, H. A. Arafat and J. H. Lienhard V, *Desalination*, 2015, **356**, 294-313.
 101. N. Sun, Z. Zhu and G. Zeng, *Sci. Total Environ.*, 2020, **744**, 140822.
 102. J. Israelachvili and R. Pashley, *Nature*, 1982, **300**, 341-342.
 103. Y.-H. Tsao, D. F. Evans and H. Wennerstrom, *Science*, 1993, **262**, 547-550.
 104. E. E. Meyer, K. J. Rosenberg and J. Israelachvili, *Proc. Natl. Acad. Sci. U. S. A.*, 2006, **103**, 15739-15746.
 105. X. An, Z. Liu and Y. Hu, *Desalination*, 2018, **432**, 23-31.
 106. C. Boo, S. Hong and M. Elimelech, *Environ. Sci. Technol.*, 2018, **52**, 14198-14207.
 107. J. Lee, C. Boo, W. H. Ryu, A. D. Taylor and M. Elimelech, *ACS Appl. Mater. Interfaces*, 2016, **8**, 11154-11161.
 108. Y. Li, Z. Zhu, J. Yu and B. Ding, *ACS Appl. Mater. Interfaces*, 2015, **7**, 13538-13546.
 109. M. Pepicelli, T. Verwijlen, T. A. Tervoort and J. Vermant, *Soft Matter*, 2017, **13**, 5977-5990.
 110. H. Liu and G. Cao, *Sci. Rep.*, 2016, **6**, 23936.
 111. S. Lin, S. Nejati, C. Boo, Y. Hu, C. O. Osuji and M. Elimelech, *Environ. Sci. Technol. Lett.*, 2014, **1**, 443-447.
 112. J. Sheng, Y. Xu, J. Yu and B. Ding, *ACS Appl. Mater. Interfaces*, 2017, **9**, 15139-15147.
 113. W. Zhang, Z. Shi, F. Zhang, X. Liu, J. Jin and L. Jiang, *Adv. Mater.*, 2013, **25**, 2071-2076.
 114. E. J. Lee, B. J. Deka, J. Guo, Y. C. Woo, H. K. Shon and A. K. An, *Environ. Sci. Technol.*, 2017, **51**, 10117-10126.
 115. L. Feng, Z. Yang, J. Zhai, Y. Song, B. Liu, Y. Ma, Z. Yang, L. Jiang and D. Zhu, *Angew. Chem.*, 2003, **115**, 4349-4352.
 116. J. Zhang and S. Seeger, *Adv. Funct. Mater.*, 2011, **21**, 4699-4704.
 117. J. Yong, Y. Fang, F. Chen, J. Huo, Q. Yang, H. Bian, G. Du and X. Hou, *Appl. Surf. Sci.*, 2016, **389**, 1148-1155.
 118. J. Zhang, J. Li and Y. Han, *Macromol. Rapid Commun.*, 2004, **25**, 1105-1108.
 119. C. Boo, J. Lee and M. Elimelech, *Environ. Sci. Technol.*, 2016, **50**, 12275-12282.
 120. V. Karanikola, C. Boo, J. Rolf and M. Elimelech, *Environ. Sci. Technol.*, 2018, **52**, 14362-14370.
 121. K. J. Lu, J. Zuo, J. Chang, H. N. Kuan and T. S. Chung, *Environ. Sci. Technol.*, 2018, **52**, 4472-4480.
 122. A. Tuteja, W. Choi, M. Ma, J. M. Mabry, S. A. Mazzella, G. C. Rutledge, G. H. McKinley and R. E. Cohen, *Science*, 2007, **318**, 1618-1622.
 123. S. Pan, A. K. Kota, J. M. Mabry and A. Tuteja, *J. Am. Chem. Soc.*, 2013, **135**, 578-581.
 124. S. S. Chhatre, W. Choi, A. Tuteja, K. C. Park, J. M. Mabry, G. H. McKinley and R. E. Cohen, *Langmuir*, 2010, **26**, 4027-4035.
 125. M. Jin, J. Wang, X. Yao, M. Liao, Y. Zhao and L. Jiang, *Adv. Mater.*, 2011, **23**, 2861-2864.
 126. J. Zhang and S. Seeger, *Angew. Chem., Int. Ed. Engl.*, 2011, **50**, 6652-6656.
 127. M. Zhang, T. Zhang and T. Cui, *Langmuir*, 2011, **27**, 9295-9301.
 128. H. Zhou, H. Wang, H. Niu and T. Lin, *Sci. Rep.*, 2013, **3**, 2964.
 129. L. Yuan, T. Wu, W. Zhang, S. Ling, R. Xiang, X. Gui, Y. Zhua and Z. Tang, *J. Mater. Chem. A*, 2014, **2**, 6952-6959.

130. H. Zhao and K. Y. Law, *Langmuir*, 2012, **28**, 11812-11818.
131. W. S. Wong, G. Liu and A. Tricoli, *Small*, 2017, **13**, 1603688.
132. W. S. Wong, G. Liu, N. Nasiri, C. Hao, Z. Wang and A. Tricoli, *ACS Nano*, 2017, **11**, 587-596.
133. B. Li and J. Zhang, *Chemical Communication*, 2016, **52**, 2744-2747.
134. H. Wang, H. Zhou, A. Gestos, J. Fang and T. Lin, *ACS Appl. Mater. Interfaces*, 2013, **5**, 10221-10226.
135. X. Li, X. Min, J. Li, N. Xu, P. Zhu, B. Zhu, S. Zhu and J. Zhu, *Joule*, 2018, **2**, 2477-2484.
136. Y. X. Huang, Z. Wang, J. Jin and S. Lin, *Environ. Sci. Technol.*, 2017, **51**, 13304-13310.
137. G. Li, Y. Lu, P. Wu, Z. Zhang, J. Li, W. Zhu, Y. Hu, D. Wu and J. Chu, *J. Mater. Chem. A*, 2015, **3**, 18675-18683.
138. Z. Wang, X. Jiang, X. Cheng, C. H. Lau and L. Shao, *ACS Appl. Mater. Interfaces*, 2015, **7**, 9534-9545.
139. M. Liu, S. Wang, Z. Wei, Y. Song and L. Jiang, *Adv. Mater.*, 2009, **21**, 665-669.
140. X. Liu, J. Zhou, Z. Xue, J. Gao, J. Meng, S. Wang and L. Jiang, *Adv. Mater.*, 2012, **24**, 3401-3405.
141. N. Wang, Y. Zhai, Y. Yang, X. Yang and Z. Zhu, *Chem. Eng. J.*, 2018, **354**, 463-472.
142. Z. Zhu, W. Wang, D. Qi, Y. Luo, Y. Liu, Y. Xu, F. Cui, C. Wang and X. Chen, *Adv. Mater.*, 2018, **30**, e1801870.
143. Z. Zhu, Z. Li, L. Zhong, R. Zhang, F. Cui and W. Wang, *J. Membr. Sci.*, 2019, **572**, 73-81.
144. A. B. D. Cassie and S. Baxter, *Trans. Faraday Soc.*, 1944, **40**, 546-551.
145. Y. Huang, Z. Wang, D. Hou and S. Lin, *J. Membr. Sci.*, 2017, **531**, 122-128.
146. T. Zhou, Y. Yao, R. Xiang and Y. Wu, *J. Membr. Sci.*, 2014, **453**, 402-408.
147. M. Q. Yang, M. Gao, M. Hong and G. W. Ho, *Adv. Mater.*, 2018, **30**, e1802894.
148. H. C. Yang, J. Hou, V. Chen and Z. K. Xu, *Angew. Chem., Int. Ed. Engl.*, 2016, **55**, 13398-13407.
149. H. Yang, W. Zhong, J. Hou, V. Chen and Z. Xu, *J. Membr. Sci.*, 2017, **523**, 1-7.
150. F. Qu, Z. Yan, H. Yu, G. Fan, H. Pang, H. Rong and J. He, *Desalination*, 2020, **486**, 114493.
151. F. Kiefer, A. Präbst, K. S. Rodewald and T. Sattelmayer, *J. Membr. Sci.*, 2019, **590**, 117294.
152. Z. Zhu, L. Zhong, T. Horseman, Z. Liu, G. Zeng, Z. Li, S. Lin and W. Wang, *J. Membr. Sci.*, 2020, DOI: 10.1016/j.memsci.2020.118768.
153. Z. Xiao, R. Zheng, Y. Liu, H. He, X. Yuan, Y. Ji, D. Li, H. Yin, Y. Zhang, X. M. Li and T. He, *Water Res.*, 2019, **155**, 152-161.
154. Z. Xiao, Z. Li, H. Guo, Y. Liu, Y. Wang, H. Yin, X. Li, J. Song, L. D. Nghiem and T. He, *Desalination*, 2019, **466**, 36-43.
155. C. Su, T. Horseman, H. Cao, K. Christie, Y. Li and S. Lin, *Environ. Sci. Technol.*, 2019, **53**, 11801-11809.
156. T. Horseman, C. Su, K. S. Christie and S. Lin, *Environ. Sci. Technol. Lett.*, 2019, **6**, 423-429.
157. Y. Ye, S. Yu, L. Hou, B. Liu, Q. Xia, G. Liu and P. Li, *Water Res.*, 2019, **149**, 588-595.
158. Z. Wang, Y. Liu, P. Tao, Q. Shen, N. Yi, F. Zhang, Q. Liu, C. Song, D. Zhang, W. Shang and T. Deng, *Small*, 2014, **10**, 3234-3239.
159. C. Boo and M. Elimelech, *Nat. Nanotechnol.*, 2017, **12**, 501-503.
160. A. V. Dudchenko, C. Chen, A. Cardenas, J. Rolf and D. Jassby, *Nat. Nanotechnol.*, 2017, **12**, 557-563.
161. R. Gupta, K. D. Rao, S. Kiruthika and G. U. Kulkarni, *ACS Appl. Mater. Interfaces*, 2016, **8**, 12559-12575.
162. Z. Deng, J. Zhou, L. Miao, C. Liu, Y. Peng, L. Sun and S. Tanemura, *J. Mater. Chem. A*, 2017, **5**, 7691-7709.
163. P. Wang, *Environ. Sci.: Nano*, 2018, **5**, 1078-1089.
164. L. Zhang, B. Tang, J. Wu, R. Li and P. Wang, *Adv. Mater.*, 2015, **27**, 4889-4894.
165. A. Politano, P. Argurio, G. Di Profio, V. Sanna, A. Cupolillo, S. Chakraborty, H. A. Arafat and E. Curcio, *Adv. Mater.*, 2017, **29**.
166. M. Fujiwara and T. Imura, *ACS Nano*, 2015, **9**, 5705-5712.
167. M. Fujiwara and M. Kikuchi, *Water Res.*, 2017, **127**, 96-103.
168. M. Fujiwara, *Desalination*, 2017, **404**, 79-86.
169. J. Wu, K. R. Zodrow, P. B. Szemraj and Q. Li, *J. Mater. Chem. A*, 2017, **5**, 23712-23719.
170. M. Ye, J. Jia, Z. Wu, C. Qian, R. Chen, P. G. O'Brien, W. Sun, Y. Dong and G. A. Ozin, *Adv. Energy Mater.*, 2017, **7**, 1601811.
171. L. Cui, P. Zhang, Y. Xiao, Y. Liang, H. Liang, Z. Cheng and L. Qu, *Adv. Mater.*, 2018, **30**, e1706805.
172. P. Tao, G. Ni, C. Song, W. Shang, J. Wu, J. Zhu, G. Chen and T. Deng, *Nat. Energy*, 2018, **3**, 1031-1041.
173. A. Ahsan, M. Imteaz, U. A. Thomas, M. Azmi, A. Rahman and N. N. Nik Daud, *Appl. Energy*, 2014, **114**, 924-930.
174. S. W. Sharshir, A. H. Elsheikh, G. Peng, N. Yang, M. O. A. El-Samadony and A. E. Kabeel, *Renewable Sustainable Energy Rev.*, 2017, **73**, 521-544.
175. G. Liu, T. Chen, J. Xu, G. Li and K. Wang, *J. Mater. Chem. A*, 2020, **8**, 513-531.
176. Y. Liu, S. Yu, R. Feng, A. Bernard, Y. Liu, Y. Zhang, H. Duan, W. Shang, P. Tao, C. Song and T. Deng, *Adv. Mater.*, 2015, **27**, 2768-2774.
177. K. Bae, G. Kang, S. K. Cho, W. Park, K. Kim and W. J. Padilla, *Nat. Commun.*, 2015, **6**, 10103.
178. W. Sun, G. Zhong, C. Kubel, A. A. Jelle, C. Qian, L. Wang, M. Ebrahimi, L. M. Reyes, A. S. Helmy and G. A. Ozin, *Angew. Chem.*, 2017, **56**, 6329-6334.
179. B. Shao, Y. Wang, X. Wu, Y. Lu, X. Yang, G. Y. Chen, G. Owens and H. Xu, *J. Mater. Chem. A*, 2020, **8**, 11665-11673.
180. Q. Zhao, N. Chen, D. Zhao and X. Lu, *ACS Appl. Mater. Interfaces*, 2013, **5**, 11453-11461.
181. Z. Liu, H. Song, D. Ji, C. Li, A. Cheney, Y. Liu, N. Zhang, X. Zeng, B. Chen, J. Gao, Y. Li, X. Liu, D. Aga, S. Jiang, Z. Yu and Q. Gan, *Glob. Chall.*, 2017, **1**, 1600003.
182. Y. Li, X. Cui, M. Zhao, Y. Xu, L. Chen, Z. Cao, S. Yang and Y. Wang, *J. Mater. Chem. A*, 2019, **7**, 704-710.
183. Y. Fu, T. Mei, G. Wang, A. Guo, G. Dai, S. Wang, J. Wang, J. Li and X. Wang, *Appl. Therm. Eng.*, 2017, **114**, 961-968.
184. A. Guo, X. Ming, Y. Fu, G. Wang and X. Wang, *ACS Appl. Mater. Interfaces*, 2017, **9**, 29958-29964.
185. A. Pal, G. Natu, K. Ahmad and A. Chattopadhyay, *J. Mater. Chem. A*, 2018, **6**, 4111-4118.
186. P. Liu, L. Miao, Z. Deng, J. Zhou, H. Su, L. Sun, S. Tanemura, W. Cao, F. Jiang and L. Zhao, *Mater. Today Energy*, 2018, **8**, 166-173.
187. M. W. Higgins, A. R. Shakeel Rahman, R. R. Devarapalli, M. V. Shelke and N. Jha, *Solar Energy*, 2018, **159**, 800-810.
188. H. Li, Y. He, Y. Hu and X. Wang, *ACS Appl. Mater. Interfaces*, 2018, **10**, 9362-9368.
189. G. Liu, J. Xu and K. Wang, *Nano Energy*, 2017, **41**, 269-284.
190. C. Wang, Y. Wang, X. Song, M. Huang and H. Jiang, *Adv. Sustainable Syst.*, 2019, **3**, 1800108.
191. L. Zhu, M. Gao, C. K. N. Peh and G. W. Ho, *Mater. Horiz.*, 2018, **5**, 323-343.
192. X. Wu, G. Y. Chen, W. Zhang, X. Liu and H. Xu, *Adv. Sustainable Syst.*, 2017, **1**, 1700046.
193. Q. Jiang, H. Gholami Derami, D. Ghim, S. Cao, Y.-S. Jun and S.

- Singamaneni, *J. Mater. Chem. A*, 2017, **5**, 18397-18402.
194. J. Yang, Y. Pang, W. Huang, S. K. Shaw, J. Schiffbauer, M. A. Pillers, X. Mu, S. Luo, T. Zhang, Y. Huang, G. Li, S. Ptasińska, M. Lieberman and T. Luo, *ACS Nano*, 2017, **11**, 5510-5518.
 195. M. Keshavarz Hedayati and M. Elbahri, *Materials (Basel)*, 2016, **9**, 497.
 196. J. Cai and L. Qi, *Mater. Horiz.*, 2015, **2**, 37-53.
 197. M. Zhu, Y. Li, F. Chen, X. Zhu, J. Dai, Y. Li, Z. Yang, X. Yan, J. Song, Y. Wang, E. Hitz, W. Luo, M. Lu, B. Yang and L. Hu, *Adv. Energy Mater.*, 2018, **8**, 1701028.
 198. G. Wang, Y. Fu, A. Guo, T. Mei, J. Wang, J. Li and X. Wang, *Chem. Mater.*, 2017, **29**, 5629-5635.
 199. L. Zhang, R. Li, B. Tang and P. Wang, *Nanoscale*, 2016, **8**, 14600-14607.
 200. J. Wang, Y. Li, L. Deng, N. Wei, Y. Weng, S. Dong, D. Qi, J. Qiu, X. Chen and T. Wu, *Adv. Mater.*, 2017, **29**, 1603730.
 201. L. Zhou, Y. Tan, J. Wang, W. Xu, Y. Yuan, W. Cai, S. Zhu and J. Zhu, *Nat. Photonics*, 2016, **10**, 393-398.
 202. Y. Xu, J. Ma, D. Liu, H. Xu, F. Cui and W. Wang, *Chem. Eng. J.*, 2019, **356**, 869-876.
 203. C. Liu, J. Huang, C.-E. Hsiung, Y. Tian, J. Wang, Y. Han and A. Fratalocchi, *Adv. Sustainable Syst.*, 2017, **1**, 1600013.
 204. X. Li, R. Lin, G. Ni, N. Xu, X. Hu, B. Zhu, G. Lv, J. Li, S. Zhu and J. Zhu, *Natl. Sci. Rev.*, 2018, **5**, 70-77.
 205. H. Ghasemi, G. Ni, A. M. Marconnet, J. Loomis, S. Yerci, N. Miljkovic and G. Chen, *Nat. Commun.*, 2014, **5**, 4449.
 206. Y. Wang, L. Zhang and P. Wang, *ACS Sustainable Chem. Eng.*, 2016, **4**, 1223-1230.
 207. M. Chen, Y. Wu, W. Song, Y. Mo, X. Lin, Q. He and B. Guo, *Nanoscale*, 2018, **10**, 6186-6193.
 208. C. Chen, Y. Li, J. Song, Z. Yang, Y. Kuang, E. Hitz, C. Jia, A. Gong, F. Jiang, J. Y. Zhu, B. Yang, J. Xie and L. Hu, *Adv. Mater.*, 2017, **29**, 1701756.
 209. Q. Jiang, L. Tian, K. K. Liu, S. Tadepalli, R. Raliya, P. Biswas, R. R. Naik and S. Singamaneni, *Adv. Mater.*, 2016, **28**, 9400-9407.
 210. L. Tian, J. Luan, K. K. Liu, Q. Jiang, S. Tadepalli, M. K. Gupta, R. R. Naik and S. Singamaneni, *Nano Lett.*, 2016, **16**, 609-616.
 211. Y. Fu, G. Wang, X. Ming, X. Liu, B. Hou, T. Mei, J. Li, J. Wang and X. Wang, *Carbon*, 2018, **130**, 250-256.
 212. J. Lou, Y. Liu, Z. Wang, D. Zhao, C. Song, J. Wu, N. Dasgupta, W. Zhang, D. Zhang, P. Tao, W. Shang and T. Deng, *ACS Appl. Mater. Interfaces*, 2016, **8**, 14628-14636.
 213. X. Hu, W. Xu, L. Zhou, Y. Tan, Y. Wang, S. Zhu and J. Zhu, *Adv. Mater.*, 2017, **29**, 1604031.
 214. W. Zhang, G. Zhang, Q. Ji, H. Liu, R. Liu and J. Qu, *ACS Appl. Mater. Interfaces*, 2019, **11**, 9974-9983.
 215. P. Mu, Z. Zhang, W. Bai, J. He, H. Sun, Z. Zhu, W. Liang and A. Li, *Adv. Energy Mater.*, 2019, **9**, 1802158.
 216. Y. Xu, C. Tang, J. Ma, D. Liu, D. Qi, S. You, F. Cui, Y. Wei and W. Wang, *Environ. Sci. Technol.*, 2020, **54**, 5150-5158.
 217. N. Xu, X. Hu, W. Xu, X. Li, L. Zhou, S. Zhu and J. Zhu, *Adv. Mater.*, 2017, **29**, 201606762.
 218. X. Li, W. Xu, M. Tang, L. Zhou, B. Zhu, S. Zhu and J. Zhu, *Proc. Natl. Acad. Sci. U. S. A.*, 2016, **113**, 13953-13958.
 219. J. Fang, Q. Liu, W. Zhang, J. Gu, Y. Su, H. Su, C. Guo and D. Zhang, *J. Mater. Chem. A*, 2017, **5**, 17817-17821.
 220. Y. Xu, H. Xu, Z. Zhu, H. Hou, J. Zuo, F. Cui, D. Liu and W. Wang, *J. Mater. Chem. A*, 2019, **7**, 22296-22306.
 221. Y. Xu, D. Liu, H. Xiang, S. Ren, Z. Zhu, D. Liu, H. Xu, F. Cui and W. Wang, *J. Membr. Sci.*, 2019, **586**, 222-230.
 222. F. Zhao, X. Zhou, Y. Shi, X. Qian, M. Alexander, X. Zhao, S. Mendez, R. Yang, L. Qu and G. Yu, *Nat. Nanotechnol.*, 2018, **13**, 489-495.
 223. X. Zhou, F. Zhao, Y. Guo, Y. Zhang and G. Yu, *Energy Environ. Sci.*, 2018, **11**, 1985-1992.
 224. X. Zhou, F. Zhao, Y. Guo, B. Rosenberger and G. Yu, *Sci. Adv.*, 2019, **5**, eaaw5484.
 225. Y. Guo, H. Lu, F. Zhao, X. Zhou, W. Shi and G. Yu, *Adv. Mater.*, 2020, **32**, e1907061.
 226. Q. Chen, Z. Pei, Y. Xu, Z. Li, Y. Yang, Y. Wei and Y. Ji, *Chemical Science*, 2018, **9**, 623-628.
 227. L. Zhu, T. Ding, M. Gao, C. K. N. Peh and G. W. Ho, *Adv. Energy Mater.*, 2019, **9**, 1900250.
 228. C. Finnerty, L. Zhang, D. L. Sedlak, K. L. Nelson and B. Mi, *Environ. Sci. Technol.*, 2017, **51**, 11701-11709.
 229. P. Zhang, J. Li, L. Lv, Y. Zhao and L. Qu, *ACS Nano*, 2017, **11**, 5087-5093.
 230. P. Mu, W. Bai, Y. Fan, Z. Zhang, H. Sun, Z. Zhu, W. Liang and A. Li, *J. Mater. Chem. A*, 2019, **7**, 9673-9679.
 231. Y. Xu, J. Ma, Y. Han, H. Xu, Y. Wang, D. Qi and W. Wang, *Chem. Eng. J.*, 2020, **384**, 123379.
 232. Z. Sun, J. Wang, Q. Wu, Z. Wang, Z. Wang, J. Sun and C. J. Liu, *Adv. Funct. Mater.*, 2019, **29**, 1901312.
 233. X. Zhou, Y. Guo, F. Zhao and G. Yu, *Acc. Chem. Res.*, 2019, **52**, 3244-3253.
 234. Y. Guo, X. Zhou, F. Zhao, J. Bae, B. Rosenberger and G. Yu, *ACS Nano*, 2019, **13**, 7913-7919.
 235. L. Zhu, M. Gao, C. K. N. Peh, X. Wang and G. W. Ho, *Adv. Energy Mater.*, 2018, **8**, 1702149.
 236. G. Xue, Y. Xu, T. Ding, J. Li, J. Yin, W. Fei, Y. Cao, J. Yu, L. Yuan, L. Gong, J. Chen, S. Deng, J. Zhou and W. Guo, *Nat. Nanotechnol.*, 2017, **12**, 317-321.
 237. H. Song, Y. Liu, Z. Liu, M. H. Singer, C. Li, A. R. Cheney, D. Ji, L. Zhou, N. Zhang, X. Zeng, Z. Bei, Z. Yu, S. Jiang and Q. Gan, *Adv. Sci.*, 2018, **5**, 1800222.
 238. X. Li, J. Li, J. Lu, N. Xu, C. Chen, X. Min, B. Zhu, H. Li, L. Zhou, S. Zhu, T. Zhang and J. Zhu, *Joule*, 2018, **2**, 1331-1338.
 239. Y. Shi, R. Li, Y. Jin, S. Zhuo, L. Shi, J. Chang, S. Hong, K.-C. Ng and P. Wang, *Joule*, 2018, **2**, 1171-1186.
 240. J. Ma, Y. Han, Y. Xu, T. Zhang, J. Zhang, D. Qi, D. Liu and W. Wang, *J. Mater. Chem. A*, 2020, DOI: 10.1039/d0ta07952a.
 241. J. Xu, Z. Wang, C. Chang, B. Fu, P. Tao, C. Song, W. Shang and T. Deng, *Desalination*, 2020, **484**.
 242. N. Xu, J. Li, Y. Wang, C. Fang, X. Li, Y. Wang, L. Zhou, B. Zhu, Z. Wu, S. Zhu and J. Zhu, *Sci. Adv.*, 2019, **5**, eaaw7013.
 243. J. Li, X. Zhou, J. Zhang, C. Liu, F. Wang, Y. Zhao, H. Sun, Z. Zhu, W. Liang and A. Li, *ACS Applied Energy Materials*, 2020, **3**, 3024-3032.
 244. Y. Xia, Q. Hou, H. Jubaer, Y. Li, Y. Kang, S. Yuan, H. Liu, M. W. Woo, L. Zhang, L. Gao, H. Wang and X. Zhang, *Energy Environ. Sci.*, 2019, **12**, 1840-1847.
 245. Y. Shi, C. Zhang, R. Li, S. Zhuo, Y. Jin, L. Shi, S. Hong, J. Chang, C. Ong and P. Wang, *Environ. Sci. Technol.*, 2018, **52**, 11822-11830.
 246. S. He, C. Chen, Y. Kuang, R. Mi, Y. Liu, Y. Pei, W. Kong, W. Gan, H. Xie, E. Hitz, C. Jia, X. Chen, A. Gong, J. Liao, J. Li, Z. J. Ren, B. Yang, S. Das and L. Hu, *Energy Environ. Sci.*, 2019, **12**, 1558-1567.
 247. Y. Kuang, C. Chen, S. He, E. M. Hitz, Y. Wang, W. Gan, R. Mi and L. Hu, *Adv. Mater.*, 2019, **31**, e1900498.
 248. Q. Zhang, G. Yi, Z. Fu, H. Yu, S. Chen and X. Quan, *ACS Nano*, 2019, **13**, 13196-13207.
 249. P. Yang, K. Liu, Q. Chen, J. Li, J. Duan, G. Xue, Z. Xu, W. Xie and J. Zhou, *Energy Environ. Sci.*, 2017, **10**, 1923-1927.
 250. G. Ni, S. H. Zandavi, S. M. Javid, S. V. Boriskina, T. A. Cooper and G. Chen, *Energy Environ. Sci.*, 2018, **11**, 1510-1519.
 251. W. Xu, X. Hu, S. Zhuang, Y. Wang, X. Li, L. Zhou, S. Zhu and J. Zhu,

- Adv. Energy Mater.*, 2018, **8**, 1702884.
252. Y. Yang, H. Zhao, Z. Yin, J. Zhao, X. Yin, N. Li, D. Yin, Y. Li, B. Lei, Y. Du and W. Que, *Mater. Horiz.*, 2018, **5**, 1143-1150.
253. C. Song, D. Qi, Y. Han, Y. Xu, H. Xu, S. You, W. Wang, C. Wang, Y. Wei and J. Ma, *Environ. Sci. Technol.*, 2020, **54**, 9025-9033.
254. J. Huang, Y. He, L. Wang, Y. Huang and B. Jiang, *Energy Convers. Manage.*, 2017, **132**, 452-459.
255. Y. Xu, J. Ma, Y. Han, J. Zhang, F. Cui, Y. Zhao, X. Li and W. Wang, *ACS Sustainable Chem. Eng.*, 2019, **7**, 5476-5485.
256. D. Hao, Y. Yang, B. Xu and Z. Cai, *ACS Sustainable Chem. Eng.*, 2018, **6**, 10789-10797.
257. L. Shi, Y. He, X. Wang and Y. Hu, *Energy Convers. Manage.*, 2018, **171**, 272-278.
258. L. Shi, Y. Shi, S. Zhuo, C. Zhang, Y. Aldrees, S. Aleid and P. Wang, *Nano Energy*, 2019, **60**, 222-230.
259. P. Qiao, J. Wu, H. Li, Y. Xu, L. Ren, K. Lin and W. Zhou, *ACS Appl. Mater. Interfaces*, 2019, **11**, 7066-7073.
260. D. Qi, Y. Liu, Y. Liu, Z. Liu, Y. Luo, Hongbo Xu, Xin Zhou, J. Zhang, Hui Yang, Wei Wang and X. Chen, *Adv. Mater.*, 2020, e2004401.

University of Singapore as a research fellow and UC Berkeley as a senior visiting scholar. His research interests are mainly focused on filtration membrane and nano-chemistry for water purification.

Xiaodong Chen is a Professor at Nanyang Technological University, Singapore. He received his B.S. degree (honors) in chemistry from Fuzhou University (China) in 1999, M.S. degree (honors) in physical chemistry from the Chinese Academy of Sciences in 2002, and Ph.D. degree (Summa Cum Laude) in biochemistry from University of Münster (Germany) in 2006. After working as a postdoctoral fellow at Northwestern University (USA), he started his independent research career as a Singapore National Research Foundation Fellow and Nanyang Assistant Professor at Nanyang Technological University, Singapore in 2009. He was promoted to Associate Professor with tenure in 2013 and Full Professor in 2016. His research interests include programmable materials for energy conversion, water remediation, and integrated nano-bio interfaces.

Biography

Zhigao Zhu received his B.S degree in polymer materials and engineering from Nanjing Tech University in 2012, M.S degree in materials engineering from Donghua University in 2014 and Doctor degree in 2019 from Wang's group at Harbin Institute of Technology. He is now an associate professor in Nanjing University of Science and Technology. His research interest mainly focuses on the superwetable nanofibrous membranes for oily water treatment and membrane distillation.

Ying Xu received her B.S degree in municipal engineering from North China University of Water Resources and Electric Power in 2014 and her Doctor degree in 2019 from Wang's group at Harbin Institute of Technology. She is now an assistant professor in Zhengzhou university. Her research interest mainly focuses on the synthesis of photo-thermal membranes for solar evaporation in water purification.

Yifei Luo is a Ph.D. student in Materials Science at Nanyang Technological University (NTU), Singapore, under the supervision of Prof. Xiaodong Chen. She received her B.E. (first class honors) in Materials Engineering from NTU in 2016. Her current research focus is hydrogel-based functional materials and devices for bioelectronic applications, such as flexible electrodes, sensors, and energy storage devices.

Wei Wang is a Professor in the School of Environment, Harbin Institute of Technology, China. He received B.S. degree in chemistry in 2007 and Ph.D. degree in polymer chemistry in 2012 from Jilin University (China). He started his independent research career at Harbin Institute of Technology in 2012 and was promoted to Full Professor in 2015. He used to work in Nanyang Technological